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# Participation in the Meeting

## DELEGATIONS FROM EUROPEAN MEMBER COUNTRIES

*Austria*

A. Zeller, Director, Landwirtschaftlich-chemische, Bundesversuchsanstalt in Wien, Wien II, Trunnerstrasse 1.

H. Linser, Studiengesellschaft für Atomenergie, Linz/Donau, Landstrasse 115/I.

*Belgium*

F. Lievens, Directeur Général, Ministère de l'Agriculture, Boulevard de Bulaissant 6, Brussels.

E. J. A. Gerard, Inspecteur en Chef, Ministère de la Santé Publique et de la Famille, 60 rue Ravenstein, Brussels.

E. Maes, Directeur, Laboratoire Central, Ministère des Affaires Economiques, 23 Rue Traversière, Brussels.

A. R. Deschreider, Laboratoire Central, Ministère des Affaires Economiques, 23 Rue Traversière, Brussels.

R. J. Guyaux, Conseiller vétérinaire, Ministère du Congo Belge et du Ruanda-Urundi, 7 Place Royale, Brussels.

M. de Faestraets, Directeur, Institut National pour l'Amélioration des Conserves de Légumes, Wesembeck-Oppen.

M. de Proost, Centre d'Etudes de l'Energie Nucléaire, Mol.

R. J. Kirchmann, Centre d'Etudes de l'Energie Nucléaire, Mol.

P. H. Manil, Institut Agronomique de l'Etat, Gembloux.

E. Schram, Faculté de Médecine, 115 Boulevard de Waterloo, Brusssels.

P. Jauniaux, "Refribel", 19 rue Proderode, Brussels.

*Denmark*

E. Stenberg Knudsen, The Danish Meat Research Institute, Roskilde.

E. Malling Olsen, Doctor of Veterinary Science, Veterinary Directorate, Nyropsgade 37, Copenhagen V.

A. Brynjolfsson, A.E.C. Research Establishment, Risø per Roskilde.

T. Truelsen, A.E.C. Research Establishment, Risø per Roskilde.

S. A. Andersen, Danish Technical University, Institute of Refrigeration Research, Oster Voldgade 8, Copenhagen K.

P. E. Andersen, National Council of Domestic Science and Household Economics, Amager Felledvej 56, Copenhagen 5.

H. Jørgensen, Professor of Technical Biochemistry, Royal Danish Technical University, 14 Odensgade, Copenhagen.

M. Jensen, Laboratory for Industrial Utilization of Fruit and Vegetables, Royal Danish Technical University, 14 Odensgade, Copenhagen.

*Finland*

Eino Jamalainen (Armas), Director of Plant Pathology, Department of Agricultural Research Centre, Tikkurila.

T. Salmi, Ministry of Agriculture, Ritarikatu 2, Helsinki.

L. Reinius, Ministry of Agriculture, Ritarikatu 2, Helsinki.

*France*

G. Mocquot, Directeur de la Station Centrale des Produits Animaux, C.N.R.Z., Jouy-en-Josas (S.-et-O.).

P. Douzou, Laboratoire de Subsistances de l'Armée, 6 Bd. des Invalides, Paris 7e.

C. Gross, Inspecteur-général, Service de la Répression des Fraudes, 42 bis rue de Bourgogne, Paris 7e.

J. Sirchis, C.E.A., Service de Biologie, Saclay.

J. Chanteur, Ministère de la Santé Publique, Institut National d'Hygiène, 3 rue Léon Bonnat, Paris 16e.

G. H. Zundel, Professeur, Ecole Vétérinaire, Alfort (Seine).

Madame S. Alibert, C.E.N., Boîte Postale No. 2, Saclay.

A. Bénézit, Section Technique du Froid, Ministère de l'Agriculture, 19 Avenue de Maine, Paris 15e.

X. Hays, Ministère des Armées (Inspection Technique des Subsistances), 8 Boulevard des Invalides, Paris 7e.

M. Mortreuil, O.R.S.T.O.M., 20 rue Monsieur, Paris.

P. Vidal, Conservatome, 18 rue Seguin, Lyon (Rhône).

F. Thiallet, Conservatome, 7 rue Parmentier, Neuilly (Seine).

G. Bigourdan, Conservatome, 18 rue Seguin, Lyon (Rhône).

P. Lévêque, C.E.A., 69 rue de Varenne, Paris, 7e.

*Germany*

J. KUPRIANOFF, Director, Bundesforschungsanstalt für Lebensmittelfrischhaltung, Karlsruhe.

SCHMIDT-LORENZ, Chief, Laboratorium für Mikrobiologie Bundesforschungsanstalt für Lebensmittelfrischhaltung, Karlsruhe.

WALTER FACHMANN, Bundesministerium für Ernährung, Landwirtschaft und Forsten, Bonn.

GÜNTER DUTSCHKE, Bundesministerium für Ernährung, Landwirtschaft und Forsten, Bonn.

KURT FRENSEN, Bundesministerium für Atomkernenergie und Wasserwirtschaft, Bad Godesberg.

DIETRICH MERTEN, Physikalisches Institut der Bundesforschungsanstalt für Milchwirtschaft, Kiel.

H. HILL, Director, Physiologisches Institut der Tierärztlichen Hochschule, Hannover.

WERNER HÖFNER, Agrikulturchemisches Institut der Universität Giessen, Giessen.

*Ireland*

T. J. LYNCH, Superintending Veterinary Officer, Department of Agriculture, Merrion Street, Dublin.

*Italy*

S. VISCOE, Direttore, Istituto Nazionale della Nutrizione, Città Universitaria, Roma.

A. MARIANI, Istituto Nazionale della Nutrizione, Città Universitaria, Roma.

M. A. SPADONI, Istituto Nazionale della Nutrizione, Città Universitaria, Roma.

D. MASSA, Direzione dei Servizi Veterinari, Ministero della Sanità, Piazza Dalmazia 1, Roma.

A. MONZINI, Vice-Direttore, Stazione Sperimentale del Freddo Via Celoria 2, Milano.

A. PORETTA, Comitato Nazionale per le Ricerche Nucleari, Stazione Sperimentale Conserve, Via Tanara 33, Parma.

C. BESSLER, Medico Provinciale, Ministero della Sanità, Roma.

G. DAL MONTE, Direzione Generale dell'Alimentazione, Ministero Agricoltura e Foreste, Roma.

*Netherlands*

M. J. L. DOLS, General Adviser to the Minister of Agriculture, Fisheries and Food, Emmapark 1, The Hague.

D. DE ZEEUW, Principal Scientific Officer, Institute of Atomic Sciences in Agriculture, Prof. Ritzema Bosweg 32, Wageningen.

H. A. LENIGER, Professor of Food Technology, Agricultural University, Prof. Ritzema Bosweg 32, Wageningen.

W. F. VAN VLIET, Institute for Research on the Storage of Agricultural Products, Bornsesteeg 59, Wageningen.

F. E. LOOSJES, Plant Protection Service, Oude Bennekomseweg 9, Wageningen.

F. F. LEUPEN, Liaison Officer for Atomic Questions, Ministry of Agriculture, Fisheries and Food, le v.d. Boschstraat 4, The Hague.

H. VAN GENDEREN, Head, Pharmaceutical and Endocrinological Laboratory, National Institute of Public Health, Zeestraat 73, The Hague.

M. VAN EEKELEN, President, Organization for Nutrition and Food Research—T.N.O., Koningskade 12, The Hague.

D. A. A. MOSSEL, Head, Laboratory of Microbiology, Central Institute for Nutrition and Food Research—T.N.O., C.I.V.O., Catherijnesingel 61, Utrecht.

*Advisers*

H. JULIUS, Professor of Hygiene, State University, Utrecht, Adviser to Zwanenberg's Meat Factories Ltd., Oss.

J. VOOGD, Senior Physicist, Philip's Research Laboratories, Eindhoven.

A. C. VAN DORSTEN, Senior Physicist, Philips' Research Laboratories, Eindhoven.

*Norway*

STEINAR HAUGE, The Veterinary College of Norway, Oslo.

KNUT MIKAELSEN, Research Director, The Agricultural College of Norway, Institute of Genetics, Vollebekk.

EIRIK HEEN, Director, Norwegian Fisheries Research Institute, P.O. Box, 187, Bergen.

ARVID EVJEN, Chief Physician, Norwegian Public Health Services, 41 Lancaster Gate, London, W.2.

*Poland*

E. PIJANOWSKI, Central College of Agriculture, ul. Rakowiecka 8, Warsaw 12.

W. BEDNARCZYK, Institute of Dairy Industries, ul. Hoza 66/68, Warsaw.

G. MAJEWSKI, St. Peter Str. 4, Gdynia.

*Portugal*

JOSÉ CORREIA CONTREIRAS, Estação Agronómica Nacional, Sacavém.

JOSÉ MANUEL ARRIAGA e CUNHA, Estação Agronómica Nacional Sacavém.

JOSÉ AMARAL MENDES, Laboratorio Central de Patologia Veterinaria, Lisbon.

M. A. DA SILVA PATO, Junta Nacional do Vinho, Lisbon.

A. M. CAETANO NUNES, Junta Nacional do Vinho, Lisbon.

R. G. FERRÃO, Junta Nacional dos Prodotos Pecuarios, Ministério de Economia, Lisbon.

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*Spain*

G. Bornás, Sub-director, Instituto Nacional de Investigaciones Agronómicas, Puerta de Hierro, Madrid.

M. C. Beltrán, Asociación de Técnicos Bromatólogos, Facultad de Farmacia, Ciudad Universitaria, Madrid.

R. Beltrán Vivar, Instituto Nacional de Industria, Plaza de Salamanca, Madrid.

J. Baumann, Instituto Nacional de Industria, Plaza de Salamanca, Madrid.

R. Lengarán, Comisaría General de Abastecimientos y Transportes Almagro 33, Madrid.

L. Mellado, Instituto Nacional de Investigaciones Agronómicas, Puerta de Hierro, Madrid.

J. A. Muñoz Delgado, Centro Experimental del Frío, Serrano 150, Madrid.

A. Valdecantos, Centro Experimental del Frío, Serrano 150, Madrid.

Bernardo Lafuente, Junta de Energía Nuclear, Departamento Química Vegetal, Paseo de Valencia al Mar, Valencia.

*Sweden*

H. Lundin, Royal Institute of Technology, Stockholm 70.

E. Abramson, National Institute of Public Health, Stockholm.

E. von Sydow, Swedish Institute for Food Preservation Research, S.I.J., Göteborg 5.

L. E. Ericson, Royal Institute of Technology in Stockholm, Stockholm 70.

A. H. Fredholm, Swedish Federation of Butchers, Stockholm 20.

*Switzerland*

H. Forster, Chimiste de la Ville de Zurich, Ausstellungsstr. 90, Zurich.

H. Mohler, Knorr Food Products, Thayngen.

R. H. Egli, Directeur des Laboratoires de Recherches, Association de l'Industrie Suisse des Produits Laitiers et Potages Afico S.A., Vevey.

P. Gfeller, Conserves Hero Lenzbourg, Lenzbourg.

Simone Radouco-Thomas, 44 Route des Acacias, Geneva.

Corneille Radouco-Thomas, Battelle Memorial Institute, Faculty of Medicine, University of Geneva, 44 Route des Acacias, Geneva.

K. Buob, Optingenstrasse 47, Berne.

M. Pâquet, Battelle Memorial Institute, 7 route de Drize, Geneva.

*United Kingdom*

N. C. Wright, Ministry of Agriculture, Fisheries and Food, Whitehall Place, London, S.W.1.

H. R. Barnell, Ministry of Agriculture, Fisheries and Food, Great Westminister House, Horseferry Road, London, S.W.1.

N. Uri, Ministry of Agriculture, Fisheries and Food, Experimental Factory, Greyhope Road, Aberdeen.

G. Wortley, Ministry of Agriculture, Fisheries and Food, Dean Bradley House, Horseferry Road, London, S.W.1.

E. E. Turtle, Ministry of Agriculture, Fisheries and Food, Infestation Control Division, Tolworth, Surrey.

W. Shortt, Ministry of Agriculture, Fisheries and Food, Great Westminster House, Horseferry Road, London, S.W.1.

M. K. Lloyd, Ministry of Agriculture, Fisheries and Food, Veterinary Lab., Weybridge, Surrey.

E. L. Taylor, Ministry of Agriculture, Fisheries and Food, Veterinary Lab., Weybridge, Surrey.

Miss J. Parker, Ministry of Agriculture, Fisheries and Food, Great Westminster House, Horseferry Road, London, S.W.1.

M. Ingram, Low Temperature Research Station (DSIR), Downing Street, Cambridge.

E. C. Bate-Smith, Low Temperature Research Station (DSIR), Downing Street, Cambridge.

J. Brooks, Low Temperature Research Station (DSIR), Downing Street, Cambridge.

Brian Coleby, Low Temperature Research Station (DSIR), Downing Street, Cambridge.

Miss M. J. Thornley, Low Temperature Research Station (DSIR), Downing Street, Cambridge.

R. A. Lawrie, Low Temperature Research Station (DSIR), Downing Street, Cambridge.

C. H. Lea, Low Temperature Research Station (DSIR), Downing Street, Cambridge.

W. G. Burton, Ditton Laboratory (DSIR), Larkfield, Maidstone, Kent.

J. M. Shewan, Torry Research Station, (DSIR), Torry, Aberdeen, Scotland.

T. A. Oxley, Pest Infestation Laboratory (DSIR), London Road, Slough, Bucks.

G. Ayert, Pest Infestation Laboratory (DSIR), London Road, Slough, Bucks.

A. B. Hammond, H. Q. Department of Scientific and Industrial Research, Charles House, 5–11 Regent Street, London, S.W.1.

Miss S. J. Attenborough, H. Q. Department of Scientific and Industrial Research, Charles House, 5–11 Regent Street, London, S.W.1.

S. Jefferson, Technological Irradiation Group, Isotope Division, Wantage Radiation Laboratory, Wantage, Berks.

T. H. Horne, Technological Irradiation Group, Isotope Division, Wantage Radiation Laboratory, Wantage, Berks.

B. A. Bridges, Technological Irradiation Group, Isotope Division, Wantage Radiation Laboratory, Wantage, Berks.

P. Cornwell, Technological Irradiation Group, Isotope Division, Wantage Radiation Laboratory, Wantage, Berks.

I. D. Clarke, Technological Irradiation Group, Isotope Division, Wantage Radiation Laboratory, Wantage, Berks.

G. S. Murray, Technological Irradiation Group, Isotope Division, Wantage Radiation Laboratory, Wantage, Berks.

Miss B. Freeman, Technological Irradiation Group, Isotope Division, Wantage Radiation Laboratory, Wantage, Berks.

Mrs. A. Marples, Technological Irradiation Group, Isotope Division, Wantage Radiation Laboratory, Wantage, Berks.

D. Dove, Technological Irradiation Group, Isotope Division, Wantage Radiation Laboratory, Wantage, Berks.

G. Glew, Technological Irradiation Group, Isotope Division, Wantage, Radiation Laboratory, Wantage, Berks.

J. R. Hickman, Technological Irradiation Group, Isotope Division, Wantage Radiation Laboratory, Wantage, Berks.

Miss M. M. Burt, Technological Irradiation Group, Isotope Division, Wantage Radiation Laboratory, Wantage, Berks.

F. Ley, Technological Irradiation Group, Isotope Division, Wantage Radiation Laboratory, Wantage, Berks.

T. Greenwood, Technological Irradiation Group, Isotope Division, Wantage Radiation Laboratory, Wantage, Berks.

D. B. Powell, Technological Irradiation Group, Isotope Division, Wantage Radiation Laboratory, Wantage, Berks.

H. Fazakerley, Technological Irradiation Group, Isotope Division, Wantage Radiation Laboratory, Wantage, Berks.

J. K. Wright, U.K. Atomic Energy Authority, 11 Charles Street, London, S.W.1.

J. Weiss, King's College, Durham University, Newcastle-on-Tyne.

H. Laser, University of Cambridge, (Medical Research Council).

G. O. Phillips, University of Cardiff.

E. C. Hill, University College, Newport Road, Cardiff.

W. F. R. Pover, Department of Medical Biochemistry and Pharmacology, University of Birmingham, Birmingham 15.

A. N. Duckham, University of Reading, Reading Berks.

S. K. Kon, National Institute for Research in Dairying, Shinfield, near Reading, Berks.

H. Burton, National Institute for Research in Dairying, Shinfield, near Reading, Berks.

S. Smith, H.M. Chemical Inspector of Factories, H.M. Factory Inspectorate, 19 St. James's Square, London, S.W.1.

F. Brookhouse, Technical Branch, Victualling Department, Admiralty.

Miss Erna Grossbard, Unit of Microbiology, Grassland Research Institute, Hurley, Berks.

Miss B. C. Hobbs, Central Public Health Laboratory (M.R.C.), Colindale Avenue, London N.W.9.

F. K. Banfield, British Food Manufacturing Industries Research Assocn., Leatherhead, Surrey.

K. D. Benton, British Baking Industries Research Association, Chorleywood, Herts.

E. N. Greer, The Research Association of British Flour Millers, Cereals Research Station, St. Albans, Herts.

Miss D. M. Evans, The Printing, Packaging and Allied Trades Research Association, Randalls Road, Leatherhead, Surrey.

N. R. Knowles, British Egg Marketing Board, Mercury House, 109–119 Waterloo Road, London, S.E.1.

R. S. Hannan, The Metal Box Co. Ltd., Research Department, Kendal Avenue, Acton, London, W.3.

M. Crowley-Milling, Metropolitan Vickers Electrical Co. Ltd., Trafford Park, Manchester 1.

C. W. Miller, Metropolitan Vickers Electrical Co. Ltd., Trafford Park, Manchester 1.

E. Broadbridge, Mullard Equipment Ltd., Mullard House, Torrington Place, London, W.C.1.

D. R. Perry, Mullard Equipment Ltd., Mullard Research Laboratories, Cross Oak Lane, Salfords, Redhill, Surrey.

J. Sainsbury, J. Sainsbury Ltd., Stamford House, Blackfriars, London, S.E.1.

E. F. Williams, J. Sainsbury Ltd., Stamford House, Blackfriars, London, S.E.1.

M. Liddington, J. Sainsbury Ltd., Stamford House, Blackfriars, London, S.E.1.

S. T. P. Brightwell, J. Sainsbury Ltd., Stamford House, Blackfriars, London, S.E.1.

A. T. S. Babb, Research Laboratories, J. Lyons and Co., 149 Hammersmith Road, London, W.14.

P. R. Booth, Kraft Foods Ltd., Morris House, Berkeley Square, London, W.1.

S. Back, Cross and Blackwell Ltd., Soho Square, London, W.1.

Miss E. A. Walker, Cyanamid of Great Britain Ltd., Bush House, Aldwych, London, W.C.2.

G. B. Galliver, Unilever Ltd., Food Research Department, Sharnbrook, Bedford.

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W. H. T. DAVISON, Tube Investment Research Laboratories, Hinxton, Nr. Cambridge.
D. G. LEWIS, Pfizer Ltd., Sandgate Road, Folkestone, Kent
P. G. SEARY, Produce Prepackaging Development Association Ltd., Agriculture House, Knightsbridge, London, S.W.1.

*Yugoslavia*

DARKO STOSIĆ, Federal Nuclear Energy Commission of F.P.R. of Yugoslavia, Kosančićev Venac 29, Belgrade.

## OBSERVERS FROM OTHER MEMBER COUNTRIES

*Australia*

J. J. MACFARLANE, Commonwealth Scientific and Industrial Research Organization, Australian Scientific Liaison Office, Africa House, Kingsway, London, W.C.2.

*Canada*

D. G. THACKER, Canadian Joint Staff, 66 Ennismore Gardens, London S.W.7.

*New Zealand*

B. W. COLLINS, Office of the High Commissioner for New Zealand, New Zealand Scientific Liaison Office, Africa House, Kingsway, London W.C.2.

Sir ERNEST MARSDEN, The Chester Beatty Research Institute, Royal Cancer Hospital, Fulham Road, London S.W.1.

*Pakistan*

R. V. QURESHI, Human Nutrition Research Unit, The Ridgeway, London N.W.7.

*United States of America*

H. LINEWEAVER, U.S. Department of Agriculture, Agricultural Research Service, Western Regional Research Laboratory, 800 Buchanan Street, Albany 10, California.

## OBSERVERS FROM INTERNATIONAL ORGANIZATIONS

*International Atomic Energy Agency*

HENRY SELIGMAN, Deputy Director-General, Head of the Department of Research and Isotopes, Kaerntnerring, Vienna I, Austria.
R. F. MOUTON, Division of Isotopes, Kaerntnerring, Vienna I, Austria.
Miss S. STEVENS, Division of Scientific and Technical Information, Kaerntnerring, Vienna I, Austria.

*World Health Organization*

I. NIR-GROSFELD, Food Additives, WHO, Palais des Nations, Geneva, Switzerland.

*Organization for European Economic Co-operation*

GEORGES REISCH, Division for Technical Action and Productivity, Agriculture and Food Directorate, OEEC, 2 rue André-Pascal, Paris XVIe, France.

*EURATOM—Communauté Européenne de l'Energie Atomique*

PIERRE KRUYS, Research Division, EURATOM, rue Belliard 51/5, Brussels, Belgium.
M. LAURENTE, Research Division, EURATOM, rue Belliard 51/5, Brussels, Belgium.

*Office International des Epizooties—OIE*

G. A. CLAXTON, Regional Veterinary Officer, Ministry of Agriculture, Fisheries and Food, Hook Rise, Tolworth, Surbiton, Surrey, United Kingdom.

*European and Mediterranean Plant Protection Organization*

V. E. WILKINS, Director-General, EMPPO. 142 Avenue des Champs-Elysées, Paris VIIIe, France.

*Commission Internationale des Industries Agricoles*

G. BORNÁS-URCULLU, INIA, Puerta de Hierro, Madrid.
P. NAVELLIER, 39 rue de Dantzig, Paris XVe, France.

*International Dairy Federation*

A. L. PROVAN, Milk Marketing Board, Thames-Ditton, Surrey, United Kingdom.
R. J. MACWALTER, United Dairies Ltd., Central Laboratory, Wood Lane, London, W.12, United Kingdom.

*Dairy Society International*

J. H. F. MCLACHLAN, Kraft Foods Ltd., Berkeley Square, London, W.1, United Kingdom.

*International Organization for Standardization*

S. JEFFERSON, Technological Irradiation Group, Isotope Division, Wantage Radiation Laboratory, Grove, Berks. c/o British Standards Institution, 2 Park Street, London, W.1, United Kingdom.

*Liaison Internationale des Industries de l'Alimentation*

Madame MATHIEU-BATSCH, LIDIA, 23, rue Notre-Dame des Victoires, Paris 2e, France.

*World Poultry Science Association*

D. H. SHRIMPTON, Low Temperature Research Station (DSIR) Downing Street, Cambridge, United Kingdom.

*International Association of Microbiological Societies*

M. INGRAM, Low Temperature Research Station, Downing Street, Cambridge, United Kingdom.

D. A. A. MOSSEL, Hoofd Bacteriologisch Laboratorium van het Centraal Institut veer Voedingsonderzoek—T.N.O. Catharijnesingel 61, Utrecht, Netherlands.

*International Institute of Refrigeration*

J. KUPRIANOFF, President of Commission 4 of the I.I.R., 177 Boulevard Malesherbes, Paris 17e, France.

## FAO STAFF AND CONSULTANTS

R. A. SILOW, Chief, Atomic Energy Branch, FAO, Rome.

N. E. HOLMES, Senior Food Technologist, Nutrition Division, FAO, Rome.

R. KREUZER, Chief, Processing Section, Fisheries Division, FAO, Rome.

HERMAN F. KRAYBILL, Curtiss Wright Corporation, Princeton, New Jersey, U.S.A. Consultant to FAO.

B. S. C. SCHWEIGERT, Director, Research and Education, American Meat Institute Foundation, 939 East 57th Street, Chicago 37, Illinois, U.S.A. Consultant to FAO.

GEORGE E. TRIPP, Chief, Packaging Engineering Radiation Division, Quartermaster Food and Container Institute of the Armed Forces, 1819 W. Pershing Road, Chicago 9, Illinois, U.S.A. Consultant to FAO.

## OFFICERS OF THE MEETING

The Meeting elected Dr. NORMAN C. WRIGHT (UK) as Chairman, and Dr. J. KUPRIANOFF (Germany) acted as Vice-Chairman.

Sessional chairmen were nominated for particular sessions as follows:

Fundamental mechanism of radiation effects: Dr. B. S. SCHWEIGERT (FAO).

The effect of radiation on important constituents of food: Dr. R. S. HANNAN (UK).

Nutritive value and wholesomeness: Dr. H. R. BARNELL (UK).

Microbiological aspects of food irradiation: Prof. H. LUNDIN (Sweden).

Present status of radiation preservation—meat, poultry and fish: Mr. G. WORTLEY (UK).

Present status of radiation preservation—eggs and milk: Prof. E. PIJANOWSKI (Poland).

Present status of radiation preservation—plant products: Dr. M. G. MOCQUOT (France).

Present status of radiation preservation—disinfestation and packaging considerations: Prof. H. JØRGENSEN (Denmark).

Economic considerations of food irradiation: Mr. J. K. WRIGHT (UK).

National food legislation affecting the use of irradiated foods: Prof. M. J. L. DOLS (Netherlands).

Country programmes and plans: Dr. D. MAES (Belgium).

Dr. R. A. SILOW (FAO) and Mr. N. E. HOLMES (FAO) served as Secretaries of the Meeting.

*Acknowledgements*

The Meeting expressed its appreciation to the Government of the United Kingdom, the United Kingdom Atomic Energy Authority, and the Director of the Atomic Energy Research Establishment, for the generous facilities, services and assistance made available. These included provision for a visit to the laboratories and irradiation facilities at the Wantage Radiation Laboratory of the Isotope Division of the Atomic Energy Research Establishment.

Valuable assistance in arrangements for the Meeting and in the preparation of the report was also given by Dr. B. COLEBY, Dr. T. HORNE, Mr. S. JEFFERSON, Mr. G. WORTLEY and Mr. N. FREND of the United Kingdom.

In determining the scope and preparing the agenda of the Meeting FAO had the benefit of the advice of a small group of experts which met at FAO headquarters in Rome in April 1958, and included:

(1) Dr. H. R. BARNELL, Scientific Adviser's Division, Ministry of Agriculture, Fisheries and Food, Great Westminster House, Horseferry Road, London, S.W.1., United Kingdom.

(2) Mr. S. JEFFERSON, Technological Irradiation Group, Atomic Energy Research Establishment, Harwell, Didcot, Berks, United Kingdom.

(3) Professor J. KUPRIANOFF, Federal Research Institute for Food Preservation, Karlsruhe, Kaiserstr. 12, Baden, Germany.

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(4) Dr. D. A. A. Mossel, Central Institute for Nutrition Research, Catharijnesingel 61, Utrecht, The Netherlands.
(5) Dr. Eirik Heen, Norwegian Fisheries Research Institute, P. O. Box 187, Bergen, Norway.
(6) Monsieur P. Lévêque, Section for Application of Radio-elements, Department of Physiochemistry, Atomic Energy Commission, Saclay, France.
(7) Dr. G. T. Scarscia, National Nuclear Research Council, Rome, Italy.
(8) Professor A. Mariani, National Institute for Nutrition, Rome, Italy.
(9) Dr. A. M. Paolucci, National Institute for Nutrition, Rome, Italy.
(10) Mr. B. Payne, International Atomic Energy Agency, Vienna, Austria.
(11) Dr. I. Nir-Grosfeld, Scientist in charge of Food Additives, Health Laboratory Methods Section, World Health Organization, Geneva, Switzerland.
(12) Mr. R. Thévenot, Director, Institut International du Froid, 177 Boulevard Malesherbes, Paris 17e, France.
(13) Mr. C. C. Thomson, OEEC, 2 rue André Pascal, Paris 16e, France.

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# Summary Report of the FAO European Meeting on the Use of Ionizing Radiations for Food Preservation

## INTRODUCTION

THE FAO European Meeting on the Use of Ionizing Radiations for Food Preservation, held by courtesy of the Government of the United Kingdom at the United Kingdom Atomic Energy Authority's Atomic Energy Research Establishment at Harwell, 17–21 November 1958, was attended by 176 representatives of 17 European member Governments and by 22 observers from 5 non-European member Governments and 14 international organizations, as listed earlier.

The meeting was convened in response to a recommendation of the first meeting of the FAO European Contact Group on the Uses of Isotopes and Radiation in Agricultural Research held at Wageningen, The Netherlands, in December 1956. Its purpose was to survey the present status of food irradiation, make a critical evaluation of the potentialities of this new technique for European countries, and consider the need for and possibility of organizing international co-operation in research on food irradiation in Europe.

Participation in the meeting included representatives of Ministries of Food, Agriculture and Fisheries having responsibility for food research programmes in their countries, representatives of atomic energy authorities, representatives of public health authorities, veterinarians, and specialists in the various disciplines concerned with food irradiation including food biochemistry, food microbiology, food storage entomology, radiation biochemistry, food wholesomeness and nutrition.

The survey of the present status of food irradiation was accomplished through the presentation of review papers by invited specialists from European countries and from the United States of America, in which country by far the greatest progress in investigations on the practical application of ionizing radiations to food preservation has been made. In addition many of the delegations presented papers on their country programmes and plans and on legislation covering the use of irradiated foods. The wide range of technical considerations on which evaluation of the potentialities of food irradiation was based is indicated by the six technical sessions into which the programme was organized and by the titles of the papers presented during these sessions. It will be seen that the scope of the meeting included not only the use of ionizing radiations for the treatment of foods in order to extend their storage life but also the disinfestation of foods, the suppression of sprouting in potatoes and root crops, and the inactivation of organisms undesirable from the public health standpoint.

At the opening session participants were welcomed on behalf of the Government of the United Kingdom by Dr. Norman C. Wright, Chief Scientific Adviser of the Ministry of Agriculture, Fisheries and Food, and on behalf of the Atomic Energy Authority by Dr. B. F. J. Schonland, Director of the Atomic Energy Research Establishment. The meeting was opened on behalf of the Director-General of FAO by Dr. R. A. Silow, Chief of the Atomic Energy Branch of FAO.



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## THE NATURE OF FOOD IRRADIATION TECHNIQUES

*The irradiation process*

The usual object of food irradiation is to increase the storage life of food by inactivating spoilage microbes, by killing insects infesting foods or by inhibiting sprout development in certain root crops and potatoes. Other applications include the killing of certain parasites in meat and the inactivation of certain specific food-poisoning organisms.

A particular advantage that irradiation offers is that it does not lead to any appreciable rise in temperature of the foodstuff during treatment. Thus perishable foods can be preserved essentially in their uncooked state.

For most applications care must be taken to prevent recontamination after treatment by the use of a suitable form of packaging. Packaging can be done before irradiation, and heat-sensitive packaging materials such as plastics as well as the more conventional rigid containers can be used.

*Sources and types of radiation*

For the irradiation of food, only $\gamma$-radiation from radioisotopes and high energy electrons from machines are normally considered. $\gamma$-Rays are similar to X-rays and have high penetrating power, whereas electrons with energies suitable for food treatment have a penetration limited to a few inches. The meeting was informed that, during the next few years, sufficiently large sources of either type could be made available for the irradiation of food.

## THE PRESENT STATE OF DEVELOPMENT

In Europe food irradiation is still in the stage of laboratory research, though there is also commercial interest in the possibilities of the process. In America plans and developments which are under way for the provision of pilot and demonstration plants reflect confidence in the process following upon the results so far obtained by laboratory studies.

The results of these studies in Europe and America indicate that the various applications of food irradiation can conveniently be grouped into three classes on the basis of the radiation dosage required to achieve particular objectives.

*Low dose treatments* (50,000 rads* or less)

Several applications of low dose treatments have been found feasible such as the suppression of sprouting in potatoes and root crops (5000 to 10,000 rads); the reproductive sterilization of insects in bulk-stored grain and certain packaged products (15,000–20,000 rads) and the killing of certain parasites in meat e.g. *Trichinella spiralis* in pork and *Cysticercus bovis* in beef (25,000–50,000 rads).

Some of these applications such as the suppression of sprouting in potatoes and root crops and the disinfestation of grain and packaged products are approaching the stage at which commercial exploitation might be considered, provided it is established that the treated foods are wholesome and free from harmful constituents.

*Moderate dose treatments* (50,000–1,000,000 rads)

Doses in this range prolong considerably the shelf life of several perishable foods stored at temperatures in the range of 0°–5°C, and a five-fold increase in the shelf-life of certain meat and meat products (chicken, pork, beef, bacon and sausage), fish and certain fruits and vegetables has been achieved. Even better results may be obtained when irradiation is combined with an antibiotic treatment. However, while these applications have definite promise, a considerable amount of further investigation will be needed before their value can be fully assessed.

The food poisoning organism *Salmonella*

* The rad is the unit recently defined to specify the dose of ionizing radiation in terms of the energy absorption per unit mass. It represents an energy absorption of 100 ergs/g of material.

which is frequently found in egg products, can be inactivated in frozen whole egg pulp by a dose of 500,000 rads. This process, which provides a unique method for destroying *Salmonella* in the frozen product, has also reached the stage at which commercial exploitation might be considered.

*High dose treatments* (2,000,000–5,000,000 rads)

These treatments are intended to give a sterile food in which no microbial spoilage is possible, and they could be applied beneficially to many foods. However, doses of this magnitude produce changes in the colour, flavour, odour and texture of all foods so far studied. The changes produced vary in degree for different foods, but are usually considered objectionable.

From the foregoing it is clear that between these three classes of dose treatments there are likely to be considerable differences in the times required for commercial development and application. In the case of all levels of dose treatments technical and economic problems will have to be overcome, and the wholesomeness of the treated food will have to be established before full scale commercial development can occur. Moreover, in the case of the high dose treatments necessary for sterilization, the problem of the undesirable changes that occur in foods subjected to such treatments will have to be solved before such treatments are likely to be used on a wide scale.

## PROBLEMS TO BE OVERCOME—FUTURE RESEARCH REQUIREMENTS

For all radiation processes, wholesomeness (including both nutritional adequacy and freedom from harmful constituents), legislative control, cost and ultimately public acceptance of the foods processed by irradiation are aspects that require consideration and further investigation.

Wholesomeness can be assessed only after long and extensive feeding trials, and such trials are an essential basis for legislation affecting the use of irradiated foods. Indeed in the absence of sufficient information on radiation effects on foods, prohibitive legislation against the use of irradiated foods has been introduced in some countries.

Feeding trials using irradiated foods have been in progress for several years in four countries and although the results to date have not shown any harmful effects, the trials have not yet progressed to a stage where an assurance of wholesomeness could be issued by a competent authority. Because such trials are time-consuming and expensive, it is highly desirable that the design of these trials prior to their commencement and the interpretation of the results after their completion should be endorsed by a competent authority. Preferably this would be the national authority which would ultimately be responsible for granting permission for the production and sale of irradiated foods. In this way there could be prior agreement concerning the nature of evidence that would be considered acceptable for the granting of such permission, and commercial clearance of irradiated foods would not be impeded.

Clearly there is a need to develop agreed methods for the testing of irradiated foods to establish their wholesomeness and to formulate fundamental principles governing the use of irradiation in food processing.

It has been clearly established that changes in the flavour, odour, texture and colour of irradiated foods present special problems. Changes of these kinds vary in degree from slight to considerable with different foods and, in the case of all foods, the changes become more marked with increasing doses. With fruits and vegetables, loss of texture is the most serious defect while with meat and meat products, changes in odours and flavours are the most important. When radiation doses are high marked changes in quality can occur and it seems that, even with prolonged and extensive studies, the chances of finding a successful solution to this problem are small. However, at low radiation doses, the changes in quality are much less marked and research may be expected to yield useful results within a reasonable period.

Radiation treatments which do not kill all

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microbes could result in the spoilage of food taking unusual paths with the result that consumers might not recognize the spoilage and, in such circumstances, food poisoning organisms might develop. Moreover, the possibility cannot be discounted that from microbes that survive irradiation, strains resistant to radiation might develop and proliferate. These would be major microbiological problems requiring fundamental studies for their solution.

The combination of irradiation with other treatments such as antibiotics, mild heating and refrigeration has already shown promise in certain instances and such combination processes should be more widely investigated. A better understanding of the chemical changes which still occur with these treatments might suggest ways to improve the processes.

Successful processes developed in the laboratory should be studied also at the pilot plant scale so that problems which might arise in production may be studied. This applies particularly to microbiological aspects of the process.

Workers in different countries report different responses to irradiation for the same kind of product. This illustrates the need for each country to investigate its own commodities and to search for possible applications, particularly suitable for its own conditions and its own commodities.

The amount of radiation required for "sterilization" depends upon several factors and the dose may well be different for different types of foods. There is no general agreement upon the criteria to be used in determining the dose to be applied, and therefore the dose cannot yet be stated precisely. These are problems requiring further study.

The limited effort which will be available for research on food irradiation should be co-ordinated with research on other methods of food preservation. By so doing unprofitable research on problems more readily solved by methods of food preservation other than irradiation will be avoided.

Present estimates indicate that costs of $\gamma$-rays and of high energy electrons will be comparable. The choice of source should be made on the basis of relative suitability for a specific process and, in all cases, the costs of treatment should be considered as including charges for both handling and irradiation. With low dose treatments in particular, handling costs may well be the major part of the total cost of treatment. For an irradiation process possessing definite advantages it appears unlikely that the cost of irradiation would be so high as to hinder its adoption. Careful studies of the costs of various treatments, including combination treatments, will be necessary in order to place food irradiation on a sound economic basis.

The use of radiation for food processing may be expected to meet with some resistance from the public. Any new process may encounter such consumer resistance but in the case of irradiated food the resistance may be more marked than is customary and considerable effort will be needed to overcome it. Evidence as to the wholesomeness of the foods, obtained as the result of the feeding trials mentioned earlier in this report, might well be one important factor in overcoming this consumer resistance.

## NATIONAL FOOD LEGISLATION AFFECTING THE USE OF IRRADIATED FOODS

Statements concerning legislation to cover the production and sale of irradiated foods were presented by several countries. In some countries existing legislation can be invoked to restrict or prohibit the sale of irradiated foods and in one country, Germany, the sale of irradiated foods is forbidden by law.

Various countries are considering the question of introducing special legislation to cover the production and sale of irradiated foods and suggestions for such legislation have several points in common, such as safeguards in relation to the wholesomeness of irradiated foods, and requirements that the process must have official approval or that the irradiated food must be labelled.

Although a considerable amount of thought has been given in several countries to the development of a satisfactory technical basis for food legislation covering the use of irradiated foods, it is apparent that the problem requires much further study. Moreover a common approach to the problem by European countries would greatly facilitate international trade in irradiated foods when they become available.

## COUNTRY PROGRAMMES AND PLANS

Accounts of current programmes and plans for future work on food irradiation presented by delegations showed that whilst, as had been known previously, a very substantial amount of fundamental research has been in progress in many fields in the United Kingdom for several years, there has also been lesser but gradually increasing activity in countries such as Belgium, Denmark, France, Germany, The Netherlands, Norway, Sweden and Yugoslavia.

These and several other countries were also contemplating the installation of new radiation sources and had plans for new or expanded programmes. There were indications of duplication of similar types of basic work in various countries, but at this early stage in investigations when experience must be gained and confirmation of preliminary findings is needed, such duplication is by no means to be deprecated. Furthermore repetition of similar types of work on particular types of foods characteristic of different countries is likely to be of value in identifying foods specially amenable to radiation treatment.

## THE NEED FOR INTERNATIONAL CO-OPERATION IN RESEARCH ON FOOD IRRADIATION IN EUROPE

Recognizing that interest in food irradiation is increasing rapidly in Europe and bearing in mind that initial costs of equipment and operational cost are high, and that many highly trained experts are needed in the various fields of specialization, it was considered that in this field of work international co-operation was particularly rewarding.

Various possible forms of co-operation were mentioned, and the value of fellowships for training and of research on an international basis, and of contacts between technical workers in different countries, both on an informal basis and through technical meetings or expert working groups on specific aspects of food irradiation, was emphasized.

## CONCLUSIONS AND OUTLOOK

(1) The meeting was impressed by the progress made in the past few years in research on the use of ionizing radiations for the treatment of foods in order to extend their storage life or inactivate undesirable organisms in them.

(2) The meeting believed that a few possible applications of irradiation such as the inactivation of *Salmonella* in egg products and of certain parasites in meat, the disinfestation of grain and certain packaged products, and the suppression of sprouting in potatoes and root crops, are approaching the stage at which commercial exploitation might be considered, provided that the wholesomeness of the treated foods had been assured.

(3) The meeting noted that whereas in these applications the remaining problems are mainly concerned with technical, economic or wholesomeness considerations, there are still many basic problems arising from radiation effects to be overcome before radiation treatment of foods for the extension of their storage life will be ready for widespread development and application. Moreover although promising results have been obtained by combining irradiation with other methods of treatment, the precise conditions for such treatments have still to be defined.

(4) The meeting recognized that the investigation of these problems will require

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time, intensive effort and the services of highly trained specialists in many fields, and unique methods may have to be developed for each foodstuff.

(5) The meeting considered that present indications are that treatments involving the use of sub-sterilizing doses of radiation rather than sterilizing doses are more likely to lead to practical applications in the reasonably near future.

(6) The meeting believed that a determined effort might lead to new and attractive methods of food preservation for particular foodstuffs which might have advantages over existing methods in regard to quality and extension of storage life.

(7) The meeting considered that the treatment of food with ionizing radiations poses similar problems to the addition to food of "food additives", and that the problem of food irradiation should be considered on a similar basis. One particular problem is that of formulating fundamental principles governing the use of radiation in food processing and developing appropriate methods for the testing of irradiated foods to establish their wholesomeness so that a common basis for legislation on this subject in individual countries may be established.

(8) The meeting also considered that it would be desirable to specify those processes applied to particular foods which might give rise to special microbiological problems as the result of radiation treatment, and to recommend microbiological methods of examination for the investigation of those problems. In such studies comparisons might be made with other methods of food processing.

(9) The meeting also considered that the problem of changes in quality of irradiated foods should receive particular attention.

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## RECOMMENDATIONS

The meeting considered that the potential value of this new method of food preservation justified considerable effort in research.

The meeting therefore recommended:

(1) that European governments should give serious consideration to means of encouraging and supporting research on food irradiation in all its aspects;

(2) that with a view to keeping developments in food irradiation under continuing review and providing opportunity for the development of international co-operation in research, a permanent technical working group on food irradiation be established by FAO, in co-operation as appropriate with other interested international organizations, and composed of expert representatives of such member governments as are engaged in or planning the early initiation of programmes of research and development in the preservation of foods by ionizing radiations;

(3) that in association with this general working group there should be established other technical groups as may be needed to deal with particular aspects of food irradiation; one such technical group might be created within the framework of FAO and WHO to study the problem of formulating fundamental principles governing the use of irradiation in food processing and of developing appropriate methods for the testing of irradiated foods to establish their wholesomeness so that a common basis for legislation on this subject in individual countries may be established. Other such technical groups might be formed to study microbiological and entomological aspects of food irradiation;

(4) that, since research and development in this field is costly, appropriate forms of international co-operation in research on food irradiation be established; FAO, in association with other international organizations, should accept the responsibility for negotiating when appropriate with European and other interested member governments, in order to develop suitable forms of co-operation;

(5) that amongst such forms of international co-operation particular emphasis be placed at this time on provision of fellowships and facilities on an international basis for training and research at laboratories experienced in food irradiation techniques.

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# Résumé du Rapport de la Réunion européenne sur l'Utilisation des Rayonnements ionisants pour la Préservation des Denrées alimentaires

## INTRODUCTION

La réunion européenne sur l'emploi des rayonnements ionisants pour la préservation des aliments, qui était organisée par la FAO et qui s'est tenue du 17 au 21 novembre 1958, au Centre de recherches atomiques de Harwell du Commissariat britannique à l'énergie atomique, grâce à la généreuse hospitalité du Gouvernement du Royaume-Uni, a été suivie par 176 représentants de 17 Etats Membres européens ainsi que par 22 observateurs de 5 Etats Membres extra-européens et de 14 organisations internationales. La liste des participants est déjà donnée.

La réunion avait été convoquée en exécution d'une recommandation formulée par le Groupe européen de contact sur l'utilisation des isotopes et des rayonnements dans la recherche agricole, lors de sa première session tenue en décembre 1956 à Wageningen (Pays-Bas). L'objet de la réunion était de faire le point des travaux sur l'irradiation des aliments, d'évaluer les potentialités qu'offre cette nouvelle technique pour les pays européens, enfin d'étudier la nécessité et la possibilité d'organiser la coopération internationale en matière de recherches sur l'irradiation des aliments en Europe.

Ont participé à la réunion des fonctionnaires qui, dans les Ministères nationaux de l'alimentation, de l'agriculture et des pêches, ont la responsabilité des recherches en matière d'alimentation; des représentants des services nationaux s'occupant de l'énergie atomique et de la santé publique; des vétérinaires et des spécialistes des diverses disciplines que met en jeu l'irradiation des aliments, notamment la biochimie et la microbiologie des aliments, l'entomologie des denrées emmagasinées, la biochimie des rayonnements, l'hygiène alimentaire et la nutrition.

Pour permettre à la réunion de faire le point de la situation actuelle dans le domaine de l'irradiation des aliments, des exposés ont été présentés par des spécialistes de pays européens et des Etats-Unis, ce dernier pays étant celui où les recherches sur l'application pratique des rayonnements ionisants pour la préservation des denrées alimentaires sont de loin les plus avancées. En outre, de nombreuses délégations ont exposé les programmes et les plans de leur pays, ainsi que la législation applicable à l'utilisation des aliments irradiés. On peut juger de la diversité des considérations techniques sur la base desquelles la réunion a évalué les potentialités de l'irradiation des aliments en considérant les six séances d'études qu'a comportées le programme et les titres des communications qui ont été présentées durant ces séances. La réunion avait à s'occuper non seulement du traitement des aliments par les rayonnements ionisants en vue de prolonger leur emmagasinage, mais aussi de la désinfestation des denrées, de l'inhibition de la germination des pommes de terre et des plantes-racines, et de l'inactivation des organismes indésirables sous l'angle de la santé publique.



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Au début de la séance inaugurale, le Dr. NORMAN C. WRIGHT, Conseiller scientifique principal au Ministère de l'agriculture, des pêches et des forêts et le Dr. B. F. J. SCHONLAND, Directeur du Centre de recherches atomiques, ont exprimé aux participants les souhaits de bienvenue, l'un du Gouvernement du Royaume-Uni, l'autre du Commissariat à l'énergie atomique. M. R. A. SILOW, Chef de la Sous-division de l'énergie atomique de la FAO, a ouvert la séance au nom du Directeur général.

## NATURE DES TECHNIQUES D'IRRADIATION DES ALIMENTS

*L'irradiation*

Le but le plus commun de l'irradiation est de prolonger la durée de conservation des aliments en inactivant les microbes capables de les détériorer, en tuant les parasites et insectes qui les infestent ou en empêchant la germination des pommes de terre et plantes-racines. Ce procédé peut également servir, entre autres applications, à détruire certains parasites des viandes et à inactiver les agents de certaines intoxications alimentaires.

L'irradiation a l'avantage particulier de ne pas entraîner d'élévation notable de température de l'aliment traité, qui se trouve donc conservé sans avoir été soumis à la chaleur.

Dans la plupart des applications, il faudra emballer convenablement le produit traité, afin d'éviter sa recontamination. Le produit peut être emballé avant traitement et il est possible d'utiliser non seulement les emballages rigides classiques, mais également des matériaux sensibles à la chaleur, tels que les matières plastiques.

*Sources et types de rayonnements*

Pour l'irradiation des aliments, on n'utilise normalement que le rayonnement gamma des radio-isotopes et les électrons à haute énergie produits par des machines. Les rayons-$\gamma$ sont analogues aux rayons-X et ont un pouvoir de pénétration élevé; par contre les électrons n'ont qu'une pénétration de quelques centimètres, aux doses d'énergie couvenant au traitement des aliments. Il a été indiqué à la réunion que des sources suffisantes de rayonnements de l'un et l'autre types seront disponibles au cours des quelques prochaines années pour l'irradiation des aliments.

## RÉSULTATS ACQUIS JUSQU'À PRÉSENT

En Europe, l'irradiation des aliments n'a pas dépassé le stade de la recherche en laboratoire, bien que les milieux commerciaux étudient les possibilités de cette méthode. En Amérique, les résultats obtenus jusqu'à présent en laboratoire ont été suffisamment encourageants pour que l'on entreprenne d'installer des usines-pilotes et de démonstration.

D'après les études effectuées en Europe et en Amérique, il apparaît commode de classer les diverses applications de la méthode d'irradiation en trois catégories, correspondant aux doses de rayonnements à appliquer pour obtenir des effets déterminés.

*Traitements à faibles doses* (50 000 rads* ou moins)

Diverses applications de ce type sont possibles; inhibition de la germination des pommes de terre et plantes-racines (5000 à 10 000 rads); stérilisation des insectes infestant les céréales en vrac et certains produits emballés (15 000 à 20 000 rads); destruction de certaines parasites de la viande, par exemple *Trichinella spiralis* du porc et *Cysticercus bovis* (25 000 à 50 000 rads).

Certaines de ces techniques, par exemple l'inhibition de la faculté germinative des pommes de terre et plantes-racines et la désinfestation des céréales et produits

* Le rad est une unité qui a été récemment définie pour exprimer une dose de radiation ionisante d'après l'énergie absorbée par unité de masse. Le rad représente 100 ergs absorbés par gramme de produit traité.

emballés, approchent du stade où il est permis d'en envisager l'exploitation commerciale, à condition toutefois qu'il soit possible d'acquérir la certitude que les aliments ainsi traités sont sains et sans danger pour le consommateur.

*Traitements à doses moyennes* (50 000 à 1 million de rads)

Des doses de cet ordre prolongent considérablement la durée de conservation de plusieurs denrées à des températures comprises entre 0 et 5°C. Par exemple, cette durée est quintuplée environ pour la viande et ses produits (poulet, porc, boeuf, bacon, saucisses), le poisson et certains fruits et légumes. Ces avantages sont plus grands encore si l'on combine l'irradiation avec un traitement aux antibiotiques. Cependant, bien que ces applications soient incontestablement prometteuses, il faudra encore de nombreuses recherches pour en déterminer exactement la valeur.

L'agent toxique *Salmonella*, qui se trouve fréquemment dans les produits à base d'oeuf, est inactivé dans la pâte d'oeuf entier congelé par une dose de 500 000 rads. Ce procédé est unique en ce sens qu'il permet la destruction de cet organisme dans le produit congelé et on a atteint le stade où il serait également justifié d'en envisager l'application commerciale.

*Traitements à hautes doses* (2 à 5 millions de rads)

Ces doses sont destinées à donner des aliments stériles, dans lesquels aucune détérioration microbienne ne peut se produire, et elles pourraient être appliquées avec profit à de nombreuses denrées. Toutefois, elles altèrent la couleur, le goût, l'odeur et la texture de tous les produits expérimentés jusqu'à présent. Ces altérations sont plus ou moins marquées selon l'aliment, mais elles sont jugées en général déplaisantes.

De ce qui précède, on voit clairement que la mise au point et l'exploitation commerciale demanderont des délais très différents selon la dose envisagée. Dans les trois cas, il faudra résoudre des problèmes techniques et économiques et acquérir la certitude que les aliments traités n'offrent aucun danger, avant de pouvoir passer à l'exploitation commerciale à grande échelle. Dans le cas des hautes doses que demande la stérilisation, il faudra en outre résoudre le problème que posent les altérations dues au traitement.

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## PROBLÈMES A RÉSOUDRE—NÉCÉSSITÉ DE NOUVELLES RECHERCHES

Quel que soit le procédé d'irradiation, il faudra consacrer de nouvelles recherches à un certain nombre d'aspects: salubrité du produit traité (c'est-à-dire aussi bien sa valeur nutritionnelle que l'absence d'éléments nocifs), surveillance légale, coût, et, en dernière analyse, accueil du public.

La salubrité ne pourra être évaluée qu'après des essais nombreux et prolongés, indispensables pour établir une législation sur l'emploi des aliments irradiés. De fait, en l'absence de renseignements suffisants concernant les effets des rayonnements sur les aliments, certains pays ont adopté une législation interdisant la vente des produits ainsi traités.

Des essais d'alimentation portant sur des denrées irradiées sont en cours depuis plusieurs années dans quatre pays et, bien qu'ils n'aient jusqu'à présent montré aucun effet nocif, ils n'ont pas encore atteint le point où l'autorité intéressée serait en mesure de certifier la salubrité des aliments testés. Ces expériences étant longues et coûteuses, il serait opportun que les méthodes d'expérimentation et les modalités d'interprétation des résultats soient approuvées au préalable par l'autorité nationale compétente, de préférence celle à qui il incomberait en définitive d'accorder le permis de produire et de mettre en vente des aliments irradiés. De la sorte, on réaliserait un accord préalable sur le type des preuves à fournir pour obtenir ce permis et la mise en vente des aliments irradiés ne rencontrerait pas d'obstacles.

Il est clair qu'il est nécessaire d'établir des méthodes agréées d'essai de salubrité des produits irradiés et de déterminer les principes

fondamentaux à observer en matière de traitement des denrées alimentaires par irradiation.

Il est absolument certain que les altérations de saveur, d'odeur, de texture et de couleur provoquées par l'irradiation posent des problèmes spéciaux. Ces altérations peuvent être légères ou considérables selon l'aliment traité et, dans tous les cas, elles s'accentuent à mesure que la dose augmente. En ce qui concerne les fruits et légumes, le ramollissement constitue le principal inconvénient; pour ce qui est de la viande et des produits de viande, c'est la modification du goût et de la saveur. Si l'on irradie à doses élevées, la qualité peut se trouver fortement altérée et il y a peu de chances que même des études prolongées et exhaustives permettent de surmonter cette difficulté. Avec des doses faibles, les altérations de qualité sont bien moins accentuées et on peut espérer que la recherche aboutira à d'utiles résultats dans un délai raisonnable.

Si l'on procède à une irradiation qui ne tue pas tous les microbes, il pourrait arriver que les aliments subissent des processus de détérioration inhabituels que les consommateurs pourraient donc ne pas reconnaître, auquel cas des agents toxiques risqueraient de se développer. En outre, on ne saurait écarter le danger d'apparition et de prolifération de souches résistantes aux rayonnements. Ce pourraient être là de sérieux problèmes de microbiologie, dont la solution demandera des études fondamentales.

L'association de l'irradiation à d'autres traitements (par exemple antibiotiques, léger chauffage ou réfrigération) est déjà apparue prometteuse dans certains cas et il faudrait développer les recherches sur ces procédés mixtes. Si l'on connaissait mieux les modifications chimiques qui se produisent même avec ces traitements, on pourrait discerner des moyens de les améliorer.

Les procédés satisfaisants qui ont été mis au point en laboratoire devraient être essayés en usines-pilotes, ce qui permettrait d'étudier les problèmes qui peuvent se poser dans la production en grand. Ceci s'applique en particulier aux aspects microbiologiques du procédé considéré.

Les chercheurs de différents pays signalent, pour un même type de produit, des réactions différentes à l'irradiation. Ceci montre que chaque pays doit étudier les denrées qu'il produit et rechercher des formules d'application convenant particulièrement aux conditions et aux denrées qui lui sont propres.

La dose de "stérilisation" dépend de plusieurs facteurs et il est fort possible qu'elle varie selon les types d'aliments traités. Il n'existe pas d'accord général sur les critères à suivre pour déterminer la dose à appliquer, et l'on ne peut donc encore définir celle-ci avec précision. Ces problèmes demandent également un supplément d'étude.

L'effort limité de recherche qu'il sera possible de consacrer à l'irradiation des aliments devrait être coordonné avec l'étude d'autres méthodes de préservation. Ceci éviterait de s'occuper sans profit aucun de problèmes qu'il est plus aisé de résoudre par des méthodes de préservation autres que l'irradiation.

D'après les estimations actuelles, la production de rayons-$\gamma$ et celle d'électrons à haute énergie seront de coût comparable. Le choix de la source devrait dépendre de sa supériorité relative pour l'application d'un procédé déterminé et, dans tous les cas, il faudrait comprendre dans le coût du traitement les frais de manutention aussi bien que d'irradiation. Dans les traitements à faibles doses en particulier, il est fort possible que la manutention soit le principal élément du coût. Pour un procédé d'irradiation présentant des avantages certains, il est improbable que le coût de l'irradiation soit prohibitif. Il faudra étudier attentivement le coût des divers traitements, y compris les traitements mixtes, afin de pouvoir déterminer en pleine connaissance de cause l'intérêt économique du procédé.

L'irradiation des aliments suscitera vraisemblablement une certaine résistance chez les consommateurs. Cette éventualité existe pour toute innovation, mais il se peut que, dans le cas de l'irradiation, la résistance soit anormalement forte et qu'il faille de gros efforts pour la surmonter.

A cet égard, les preuves de salubrité qui résulteraient des essais d'alimentation mentionnés précédemment pourraient constituer un important facteur.



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## LÉGISLATIONS NATIONALES TOUCHANT L'UTILISATION DES ALIMENTS IRRADIÉS

Plusieurs pays ont présenté des exposés sur leur législation en matière de production et de vente des aliments irradiés. Certains pays peuvent invoquer les textes actuels pour limiter ou empêcher la vente de ces produits et dans un cas—l'Allemagne—cette vente est interdite par la loi.

Divers pays envisagent d'instituer une législation spéciale applicable à la production et à la vente des aliments irradiés et les suggestions formulées à cet égard coincident sur plusieurs points, par exemple garanties de salubrité des produits, soumission du procédé de traitement à l'agrément officiel ou obligation d'étiqueter comme telles les denrées irradiées.

Bien que plusieurs pays aient longuement considéré la question de la mise au point d'une base technique satisfaisante sur laquelle se fonderait la législation relative à l'utilisation des aliments irradiés, il apparaît que le problème demande encore de nombreuses études. De plus, lorsque les aliments irradiés deviendront disponibles, leur commerce international serait considérablement facilité si les pays européens abordaient le problème de législation selon une formule commune.

## PROGRAMME ET PLANS NATIONAUX

D'après la description que les délégations ont donnée des programmes en cours et des plans d'activités futures de leur pays en matière d'irradiation des aliments, il apparaît que si—comme on le savait d'ailleurs—le Royaume-Uni conduit depuis plusieurs années de très importantes recherches fondamentales dans de nombreux domaines, les activités de divers pays, tels que la Belgique, le Danemark, la France, l'Allemagne, les Pays-Bas, la Norvège, la Suède et la Yougoslavie, quoique moindres, se sont graduellement intensifiées.

Ces pays et plusieurs autres envisagent également d'installer de nouvelles sources de rayonnements et ils prévoient des programmes nouveaux ou élargis. Il apparaît que plusieurs pays poursuivent en même temps des travaux identiques, mais de telles répétitions ne sont nullement inutiles car les recherches en sont encore à la phase initiale, où il faut développer l'expérience et confirmer les résultats préliminaires acquis. De plus, la répétition de travaux sur des types particuliers d'aliments caractéristiques de différents pays aidera probablement à déterminer les denrées qui se prêtent spécialement bien à l'irradiation.

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## NÉCÉSSITÉ D'UNE COOPÉRATION INTERNATIONALE EN MATIÈRE DE RECHERCHES SUR L'IRRADIATION EN EUROPE

Etant donné la rapidité avec laquelle l'intérêt à l'égard de l'irradiation des aliments se développe en Europe et les dépenses considérables qu'exigent l'achat, le fonctionnement et l'entretien de l'équipement, ainsi que le grand nombre de spécialistes très qualifiés qui sont nécessaires, la réunion estime que la coopération internationale serait particulièrement profitable.

On a mentionné les diverses formes que pourrait prendre cette coopération, en insistant sur l'intérêt des bourses internationales de perfectionnement et de recherche, et des contacts établis soit à titre non-officiel, soit au sein de réunions techniques ou de groupes d'experts s'occupant d'aspects déterminés de l'irradiation des aliments.

## CONCLUSIONS

(1) La réunion a été impressionnée par les progrès réalisés au cours des quelques dernières années par les recherches sur l'emploi des rayonnements ionisants pour le traitement des denrées alimentaires en vue de prolonger la durée de conservation de ces

produits ou d'inactiver les organismes qui les infestent.

(2) La réunion estime qu'en ce qui concerne quelques-unes des applications possibles—par exemple inactivation des *Salmonella* dans les produits à base d'oeufs et de certains parasites des viandes, désinfestation des céréales et de certains produits empaquetés, inhibition de la germination des pommes de terre et des plantes-racines—on approche du stade où il est permis d'envisager l'exploitation commerciale, à condition que soit préservée la salubrité des aliments traités.

(3) La réunion observe que, alors que dans les applications susmentionnées les difficultés qui restent à surmonter sont essentiellement d'ordre technique, économique et sanitaire, il faudra encore résoudre de nombreux problèmes fondamentaux suscités par les effets des rayonnements avant que la technique d'irradiation des aliments en vue d'en prolonger l'emmagasinage puisse être développée et appliquée largement. De plus, si la combinaison de l'irradiation avec d'autres méthodes de traitement a donné des résultats prometteurs, il reste néanmoins à définir avec précision les conditions d'emploi de ces techniques.

(4) La réunion reconnaît que l'étude de ces problèmes exigera du temps, beaucoup d'efforts et un personnel hautement spécialisé dans de nombreux domaines, et qu'il faudra peut-être mettre au point des méthodes particulières pour chaque denrée alimentaire.

(5) La réunion est d'avis que, d'après ce que l'on sait actuellement, ce sont les traitements faisant appel à des doses inférieures aux doses de stérilisation qui semblent le plus susceptibles d'application pratique dans un avenir raisonnablement proche.

(6) La réunion pense qu'un effort vigoureux pourrait aboutir à la découverte de méthodes intéressantes de préservation de certains aliments, méthodes qui, par rapport aux techniques actuelles, pourraient offrir des avantages considérables sous les aspects, qualité du produit et durée de conservation.

(7) La réunion estime que le traitement des produits alimentaires au moyen de rayonnements ionisants pose des problèmes analogues à ceux que soulève l'adjonction de certaines substances aux aliments et que la question de l'irradiation devrait être envisagée de la même manière. Un problème particulier consiste à formuler les principes de base devant régir l'emploi des rayonnements pour le traitement des aliments et à mettre au point des méthodes convenables pour le contrôle des aliments irradiés, de façon à établir une base commune sur laquelle les divers pays pourraient fonder leur législation en la matière.

(8) La réunion estime qu'il serait opportun de préciser quels sont ceux des procédés d'irradiation appliqués à des aliments particuliers qui pourraient soulever des problèmes spéciaux de microbiologie, et de recommander des méthodes d'examen microbiologique à suivre pour l'étude de ces problèmes. Au cours de telles études, on pourrait procéder à des comparaisons avec d'autres méthodes de préservation.

(9) La réunion estime enfin qu'il conviendrait d'étudier spécialement le problème de l'altération de la qualité des aliments irradiés.

## RECOMMANDATIONS

La réunion voit dans l'irradiation des denrées alimentaires un procédé de conservation riche de possibilités nouvelles. Elle recommande en conséquence:

(1) que les gouvernements des pays d'Europe étudient attentivement les moyens d'encourager et d'appuyer la recherche en matière d'irradiation des aliments sous tous ses aspects;

(2) que, afin qu'il soit possible de suivre les progrès dans le domaine de l'irradiation des aliments et de développer la coopération internationale en matière de recherche, la FAO, coopérant de la manière la plus appropriée avec les autres organisations internationales intéressées, constitue un groupe de travail technique permanent sur l'irradiation des aliments, composé d'experts

représentant les gouvernements des Etats Membres qui ont entrepris ou envisagent d'entreprendre à brève échéance des programmes de recherche et de développement en matière de préservation des aliments par les rayonnements ionisants;

(3) que soient constitués, en relation avec le groupe général de travail susmentionné, tels autres groupes qui pourraient être nécessaires pour étudier certains aspects particuliers de l'irradiation des aliments. Un de ces groupes techniques pourrait être créé dans le cadre de la FAO et de l'OMS, avec mandat d'étudier le problème que pose l'établissement de principes de base devant régir l'emploi de l'irradiation dans le traitement des produits alimentaires et la mise au point de méthodes convenables de contrôle des aliments irradiés, afin de fournir une base commune aux diverses législations nationales en la matière. Un autre groupe technique pourrait être créé pour s'occuper des aspects microbiologiques et entomologiques de l'irradiation;

(4) qu'étant donné le coût élevé des travaux de recherche et de développement dans le domaine considéré, on établisse entre les pays européens les formes appropriées de collaboration internationale. La FAO conjointement avec d'autres organisations internationales devrait, lorsqu'il y a lieu, se charger de négocier avec les gouvernements des Etats Membres européens ou autres gouvernements intéressés, en vue de définir des formes convenables de coopération;

(5) que, parmi les formes que pourrait prendre cette coopération, on s'attache tout spécialement à accorder sur une base internationale des bourses et autres moyens de perfectionnement et de recherche, dans des laboratoires possédant l'expérience des techniques d'irradiation des aliments.

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# Resumen del Informe de la Reunión Europea Sobre el Empleo de Radiaciones Ionizantes en la Preservación de Alimentos

## INTRODUCCION

A la Reunión Europea sobre el Empleo de Radiaciones Ionizantes en la Preservación de Alimentos, celebrada del 17 al 21 de noviembre de 1958, por invitación del Reino Unido, en el Establecimiento de Investigaciones sobre Energía Atómica de Harwell, dependiente de la "Atomic Energy Authority" de dicho país, asistieron 176 representantes de 17 Estados Miembros europeos y 22 observadores de cinco Estados Miembros no europeos y de 14 Organizaciones internacionales que se enumeraron previamente.

La reunión fué convocada atendiendo una recomendación formulada en la primera reunión del Grupo Europeo de Contacto sobre las Aplicaciones de las Radiaciones e Isótopos Radiactivos en las Investigaciones Agronómicas celebrada en diciembre de 1956 en Wageningen (Países Bajos), siendo su finalidad analizar el estado actual de la irradiación de alimentos, proceder a una evaluación crítica de las potencialidades de esta nueva técnica para los países europeos y examinar la necesidad y posibilidad de organizar en Europa la cooperación internacional en las investigaciones sobre irradiación de productos alimenticios.

Asistieron representantes de los Ministerios de Alimentación, Agricultura y Pesca encargados de los programas de investigaciones bromatológicas en sus respectivos países, de los centros de energía atómica, de las autoridades sanitarias, veterinarios y especialistas en las diversas materias relacionadas con la irradiación de alimentos, comprendiendo bioquímica y microbiología de los alimentos, entomología de productos alimenticios almacenados, aspectos bioquímicos de los efectos de las radiaciones, valor nutritivo e inocuidad de los productos alimenticios sometidos a ratamiento y nutrición.

El estado actual de la irradiación de alimentos fué analizado en trabajos presentados por especialistas europeos y de los Estados Unidos de América, país este que se destaca con mucho en cuanto a los progresos logrados en investigaciones sobre la aplicación práctica de las radiaciones ionizantes en la preservación de alimentos. Además, muchas de las delegaciones presentaron trabajos sobre los planes y programas de sus respectivos países y sobre la legislación referente al empleo de alimentos irradiados. De la larga serie de consideraciones técnicas en que se basó la evaluación de las potencialidades de la irradiación de alimentos son exponente las seis sesiones técnicas en que se dividió el programa y los títulos de los documentos presentados. Se advertirá que el temario de la reunión abarcaba no sólo el empleo de las radiaciones ionizantes para prolongar el almacenamiento de los productos alimenticios, sino también su desinfestación, la supresión de la germinación en las patatas y raíces y la inactivación de organismos nocivos desde el punto de vista sanitario.

En la sesión de apertura dieron la bienvenida a los participantes el Dr. Norman C. Wright, Asesor Científico Jefe del Ministerio de Agricultura, Pesca y Alimentación, del Reino Unido, en nombre del Gobierno británico, y el Dr. B. F. J. Schonland, Director del Establecimiento de Investigaciones sobre Energía Atómica, en nombre de la "Atomic Energy Authority". En nombre del Director General de la FAO declaró abierta la Reunión el Dr. R. A. Silow, Jefe del Departamento de Energía Atómica de la Organización.

## NATURALEZA DE LAS TECNICAS DE IRRADIACION DE ALIMENTOS

*El proceso de irradiación*

Por lo común, el objeto que con la irradiación de alimentos se persigue es prolongar su almacenamiento inactivando los microbios que producen su deterioro, provocar la muerte de los insectos que los infestan o inhibir la germinación en ciertas raíces y patatas. Otras aplicaciones comprenden la esterilización de determinados parásitos de la carne y la inactivación de ciertos organismos que corrompen los productos alimenticios.

Una particular ventaja que presenta la irradiación estriba en no provocar un aumento sensible de la temperatura del producto alimenticio durante el tratamiento. Así, los alimentos perecederos pueden preservarse esencialmente en su estado crudo.

En la mayor parte de las aplicaciones hay que procurar, recurriendo al oportuno procedimiento de envase, que los productos no vuelvan a contaminarse después del tratamiento. Cabe proceder al envase antes de la irradiación, pudiéndose utilizar materiales sensibles al calor, como los plásticos, y los envases rígidos corrientes.

*Fuentes y tipos de radiaciones*

Para la irradiación de los alimentos sólo se emplean, en general, rayos gamma de isótopos radiactivos y electrones de gran energía producidos por maquinaria especial. Los rayos-$\gamma$ son semejantes a los rayos-X y tienen gran poder de penetración, en tanto que el de los electrones dotados de energía apta para el tratamiento de los productos alimenticios se limita a unas cuantas pulgadas. La Conferencia tomó nota de que en el curso de los próximos años podrán habilitarse fuentes suficientemente abundantes de ambos tipos para la irradiación de alimentos.

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## ESTADO ACTUAL DE LOS TRABAJOS

En Europa, la irradiación de alimentos se encuentra todavía en la fase de laboratorio, aunque los medios industriales también se interesan por las posibilidades que entraña. En los Estados Unidos de América los planes elaborados y los trabajos en curso para la construcción de instalaciones experimentales y de demostración práctica son exponentes de las esperanzas puestas en el nuevo procedimiento en vista de los resultados hasta ahora obtenidos en los estudios de laboratorio.

Los resultados de estos estudios en Europa y América indican que las diversas aplicaciones de la irradiación de alimentos pueden agruparse en tres clases con arreglo a la dosis de radiación necesaria para alcanzar determinados objetivos.

a. *Tratamiento con dosis bajas* (máximo de 50.000 rads*)

Se ha comprobado que los tratamientos con dosis bajas son susceptibles de diversas aplicaciones como la supresión de la germinación en las patatas y en las raíces (de 5.000 a 10.000 rads); la esterilización de insectos en cereales almacenados a granel y en ciertos productos envasados (de 15.000 a 20.000 rads) y la destrucción de ciertos parásitos de la carne, v.gr. *Trichinella spiralis* en el cerdo y *Cysticercus bovis* en los bovinos (de 25.000 a 50.000 rads).

Algunas de estas aplicaciones, como la supresión de las germinación en las patatas y en las raíces cultivadas y la desinfestación de cereales y de productos envasados, van acercándose a la fase en que podría estudiarse su explotación industrial, siempre que previamente se compruebe la inocuidad de los alimentos sometidos a tratamiento y el hecho de estar éstos libres de componentes nocivos.

b. *Tratamiento con dosis medias* (de 50.000 a 1.000.000 de rads)

Las dosis comprendidas dentro de estos límites prolongan considerablemente la duración de varios productos alimenticios

* El rad es la unidad recientemente definida para designar las dosis de radiación ionizante expresada en absorción de energía por unidad de masa. Representa una absorción de energía de 100 ergios por gramo de material.

perecederos almacenados a temperaturas comprendidas entre 0° y 5°C, habiéndose conseguido aumentar en cinco veces la vida de almacenamiento de ciertas carnes y productos cárnicos (carnes de aves, cerdo, vacuno, "bacon" y salchichería), del pescado y de ciertas frutas y hortalizas. Cabe obtener resultados todavía mejores cuando la irradiación va unida a un tratamiento con antibióticos. Sin embargo, aunque estas aplicaciones infunden esperanzas que seguramente no quedarán defraudadas, será necesario llevar a cabo una considerable labor investigadora antes de que sea posible determinar plenamente el valor que revisten.

El organismo tóxico *Salmonella*, que se encuentra con frecuencia en los productos derivados de huevos, puede inactivarse en la mezcla de claras y yemas sometiéndola a una dosis de 500.000 rads. Este procedimiento, que constituye un método único de destruir *Salmonella* en el producto congelado, ha llegado también a la fase en que podría pensarse en su explotación industrial.

c. *Tratamiento con dosis altas* (de 2.000.000 a 5.000.000 de rads)

Con estos tratamientos se persigue la finalidad de obtener alimentos esterilizados en que no sea posible su corrupción microbiana; podrían aplicarse provechosamente a muchos alimentos. Sin embargo, las dosis de esta magnitud provocan cambios de color, sabor, olor y textura en todos los alimentos estudiados hasta la fecha. El grado del cambio producido varía en distintos alimentos, pero generalmente suele considerarse inaceptable.

De lo dicho anteriormente se desprende que estos tres grupos de tratamientos presentarán probablemente diferencias considerables en cuanto al espacio de tiempo que su desarrollo y aplicaciones industriales habrán de exigir. En todos ellos habrá que resolver problemas de orden técnico y económico y antes de que sea posible acometer la producción industrial en gran escala será imprescindible poner fuera de toda duda el valor nutritivo y la inocuidad de los productos sometidos a tratamiento. A mayor abundamiento, por lo que respecta a los tratamientos con las dosis elevadas necesarias para la esterilización, se impondrá resolver el problema de las alteraciones que aquéllos producen en los alimentos, antes de aplicarlos en gran escala.

## PROBLEMAS POR RESOLVER—NECESIDADES DE INVESTIGACION EN EL FUTURO

En todos los procesos de radiación, el valor nutritivo e inocuidad de los productos tratados, la reglamentación legislativa, el coste y en último término la aceptación de los productos irradiados por parte del público son aspectos que se impone examinar e investigar más detenidamente.

La inocuidad y valor nutritivo de los productos sólo pueden evaluarse después de efectuar prolongados y extensos estudios bromatológicos, constituyendo tales ensayos una base esencial para la legislación sobre el empleo de productos irradiados. De hecho, a falta de datos suficientes sobre los efectos de la irradiación en los alimentos, en algunos países está prohibido por la ley el empleo de productos alimenticios irradiados.

En cuatro países vienen efectuándose desde hace varios años ensayos de ingestión de alimentos irradiados y aunque hasta la fecha no se ha observado efecto nocivo alguno, los ensayos realizados no han alcanzado la fase en que una autoridad competente pueda garantizar la inocuidad de tales productos. Como tales ensayos requieren tiempo y resultan costosos, es sumamente conveniente que tanto su preparación como la interpretación de sus resultados lleven el refrendo de una autoridad competente. Esta debe ser preferentemente la autoridad nacional a que en última instancia correspondería conceder el permiso para la fabricación y venta de productos irradiados. De este modo podría establecerse un acuerdo previo sobre la naturaleza de los datos que se considerarían aceptables para conceder tal autorización y para que se pusieran a la venta los productos irradiados.

Es manifiesta la necesidad de elaborar métodos, aprobados de común acuerdo, para el ensayo de alimentos irradiados que permitan determinar su inocuidad y asimismo la de formular principios fundamentales que regulen el empleo de las radiaciones en la elaboración de alimentos.

Se ha puestro fuera de toda duda que los cambios de sabor, olor, textura y color de los alimentos irradiados plantean problemas especiales. Las alteraciones de esta naturaleza varían de intensidad con los distintos productos, siendo leves en unos y considerables en otros y acentuándose siempre al aumentar la dosis. Tratándose de frutas y hortalizas, la pérdida de textura constituye el defecto más grave; en la carne y en los productos cárnicos, los cambios de olor y sabor son los más importantes. Cuando las dosis de radiación son elevadas, pueden presentarse marcadas alteraciones de calidad y parece que, aun realizando estudios prolongados y extensos, las probabilidades de encontrar una solución positiva de este problema son escasas. Sin embargo, cuando las dosis de radiación son bajas, los cambios de calidad son mucho menos marcados y es de esperar que las investigaciones arrojen resultados útiles dentro de un espacio de tiempo razonable.

Los tratamientos con radiaciones que no matan todos los microbios pueden traducirse en que el deterioro de los productos tome rumbos inusitados, lo que puede tener por consecuencia que los consumidores no adviertan el deterioro, y en tales circunstancias pueden desarrollarse organismos tóxicos. Además, no cabe descartar la posibilidad de que los microbios que sobrevivan a la irradiación produzcan cepas resistentes a ésta y proliferen, planteando grandes problemas microbiológicos cuya solución exigiría investigaciones científicas puras.

La combinación de la irradiación con otros tratamientos, como el uso de antibióticos, del calentamiento suave y de la refrigeración han dado ya en algunos casos resultados prometedores, por lo que tales procedimientos mixtos deben investigarse más ampliamente. Un conocimiento más profundo de los cambios químicos que todavía se operan con estos tratamientos podría sugerir la manera de perfeccionar estos procedimientos.

Los procedimientos que han dado buen resultado en laboratorio deben estudiarse también en la instalación experimental, de modo que sea posible abordar los problemas que puedan surgir en la producción; esto se aplica particularmente a los aspectos microbiológicos.

Científicos de diversos países han comunicado distintas reacciones a la irradiación por parte de productos de la misma clase, hecho que ilustra la necesidad de que cada país investigue sus productos y estudie posibles aplicaciones que se adapten especialmente a sus condiciones y productos propios.

La cuantía de la irradiación necesaria para lograr la "esterilización" depende de diversos factores y la dosis puede variar para distintos tipos de productos alimenticios. No existe unanimidad acerca de los criterios que hay que seguir para determinar la dosis que debe aplicarse, por lo que todavía no es posible fijarla con precisión. Se trata de problemas que requieren ulterior estudio.

Los limitados esfuerzos que podrán dedicarse a investigaciones sobre la irradiación de alimentos deben coordinarse con los estudios sobre otros métodos de preservación. De ese modo se podrán evitar investigaciones poco rentables sobre problemas que pueden resolverse más fácilmente siguiendo métodos de preservación distintos de la irradiación.

Estimaciones actuales indican que los costes de los rayos-$\gamma$ y de los electrones de gran energía resultarán comparables. A la selección de la fuente debe procederse atendiendo a la idoneidad relativa para un procedimiento determinado y en todos los casos debe considerarse que los costes de tratamiento comprenden los gastos de manipulación e irradiación. En particular, en los tratamientos de dosis bajas los costes de manipulación pueden muy bien constituir la parte principal del coste total. Si se trata de un procedimiento de irradiación que presente ventajas concretas, se considera improbable que el coste de la irradiación resulte tan elevado que impida ponerla en práctica. Se impondrá efectuar estudios detenidos sobre los costes de diversos tratamientos,

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comprendidos los mixtos, a fin de asentar la irradiación de alimentos sobre una base económica sólida.

Es de suponer que el empleo de radiaciones en la elaboración de productos alimenticios tropiece con resistencia por parte del público. Todo nuevo procedimiento puede encontrar resistencia en los consumidores, pero en el caso de los productos alimenticios irradiados, tal resistencia puede ser más pronunciada de lo corriente y será necesario un considerable esfuerzo para vencerla.

Los estudios bromatológicos sobre el valor nutritivo e inocuidad de los productos que aporten los ensayos antes mencionados en este informe pueden muy bien constituir un factor importante para superar esta resistencia por parte de los consumidores.

## DISPOSICIONES LEGISLATIVAS NACIONALES QUE AFECTAN AL EMPLEO DE ALIMENTOS IRRADIADOS

Diversas delegaciones presentaron exposiciones relativas a la legislación sobre la producción y venta de productos alimenticios irradiados. En algunos países, pueden invocarse disposiciones legislativas vigentes para restringir o prohibir la venta de productos alimenticios irradiados y en uno de ellos—Alemania—está prohibida por la ley la venta de tales productos.

Diversos países tienen en estudio la cuestión relativa a la implantación de legislación especial sobre la producción y venta de alimentos irradiados y las sugerencias formuladas para tal legislación tienen varios puntos en común, como garantías en cuanto al valor nutritivo e inocuidad de los productos irradiados y el requisito de que el procedimiento ha de contar con la aprobación oficial o de que se proceda a etiquetar los alimentos irradiados.

Aunque en diversos países se ha estudiado en medida considerable la cuestión de base técnica satisfactoria para la legislación sobre alimentos que afecte también al empleo de productos irradiados, es evidente que el problema exige un estudio mucho más amplio. Además, si los países europeos abordaran al problema conjuntamente, ello facilitaría en gran manera el comercio internacional de productos alimenticios irradiados cuando se disponga de ellos.

## PROGRAMAS Y PLANES DE LOS DISTINTOS PAISES

Las exposiciones sobre los actuales planes y programas de actividades futuras sobre irradiación de alimentos hechas por las delegaciones pusieron de manifiesto que si bien, como ya era sabido, en el Reino Unido han venido llevándose a cabo durante varios años una labor muy considerable de investigación pura en diversas materias, también se han desarrollado actividades en países como Alemania, Bélgica, Dinamarca, Francia, Noruega, los Países Bajos, Suecia y Yugoeslavia, actividades de menor consideración, pero que van intensificándose gradualmente.

Estos y otros diversos países tienen asimismo en estudio la instalación de nuevas fuentes de radiación y planean programas nuevos o ampliados. Hay indicios de duplicidad de trabajos fundamentales de tipos similares en distintos países, pero en esta primera etapa de las investigaciones, en que hay que adquirir experiencia y confirmar los resultados provisionales, tal duplicación no es de lamentar en modo alguno. Además, es probable que la repetición de tipos semejantes de trabajos sobre determinados tipos de alimentos característicos de distintos países resulte útil para identificar los que se presten especialmente al tratamiento por radiaciones.

## NECESIDAD DE COOPERACION INTERNACIONAL EN LAS INVESTIGACIONES SOBRE LA IRRADIACION DE ALIMENTOS EN EUROPA

Reconociendo que el interés por la irradiación de productos alimenticios va aumentando rápidamente en Europa y teniendo en cuenta que los gastos iniciales

por equipo y el coste de funcionamiento son cuantiosos, y que se necesitan muchos expertos sólidamente capacitados en los diversos campos de especialización se consideró que la cooperación internacional es particularmente remuneradora en esta esfera de actividades.

Se hizo referencia a posibles formas de cooperación, subrayándose el valor de las becas de capacitación y de las investigaciones sobre base internacional, y del mantenimiento de contacto entre técnicos de distintos países tanto con carácter extraoficial como mediante reuniones técnicas o grupos de trabajo de expertos sobre aspectos concretos de la irradiación de alimentos.

## CONCLUSIONES Y PERSPECTIVAS

(1) La Reunión quedó favorablemente impresionada ante los progresos realizados en estos últimos años en el sector de las investigaciones sobre el tratamiento de los productos alimenticios con radiaciones ionizantes para prolongar su almacenamiento o inactivar los organismos nocivos que contienen.

(2) La Reunión estimó que unas cuantas posibles aplicaciones de la irradiación como la inactivación de *Salmonella* en los derivados de huevos y de ciertos parásitos de la carne, la desinfestación de cereales y de ciertos productos envasados y supresión de la germinación en patatas y raíces van aproximándose a la fase en que podría estudiarse su explotación industrial, siempre que se ponga fuera de toda duda la inocuidad de los productos tratados.

(3) La Reunión tomó nota de que, mientras en estas aplicaciones los problemas que quedan por resolver se refieren principalmente a los aspectos técnicos, económicos y de inocuidad, subsisten aún muchos problemas fundamentales, planteados por los efectos de las radiaciones que hay que resolver para que la irradiación de productos alimenticios encaminados a prolongar su almacenamiento pueda ser objeto de un amplio desarrollo y aplicación. Además, aunque la combinación de la irradiación con otros métodos ha dado resultados prometedores, están todavía por definir las condiciones precisas en que tales tratamientos podrán realizarse.

(4) La Reunión reconoció que el estudio de estos problemas exigirá tiempo, esfuerzo intenso y los servicios de especialistas de sólida capacitación en muchas materias, siendo posible que haya que elaborar métodos especiales para cada producto alimenticio.

(5) La Reunión consideró que en la actualidad todo induce a creer en la probabilidad de que en un futuro no muy lejano las aplicaciones prácticas se consiguen con los tratamientos que suponen el empleo de dosis de radiación subesterilizante y no de dosis esterilizantes.

(6) En la Reunión se expresó la creencia de que un esfuerzo tenaz puede traducirse en métodos nuevos e interesantes de conservación de determinados productos alimenticios, métodos que puedan ofrecer ventajas sobre los procedimientos actuales en lo que respecta a la calidad y prolongación de su almacenamiento.

(7) La Reunión estimó que el tratamiento de alimentos con radiaciones ionizantes plantea problemas análogos al de los aditivos alimentarios y que el problema de la irradiación debe plantearse sobre una base análoga. Un problema especial lo plantea la formulación de principios fundamentales que regulen el empleo de radiaciones en la elaboración de alimentos y el desarrollo de métodos adecuados para el ensayo de productos irradiados a fin de determinar su inocuidad de modo que pueda establecerse una base común para la legislación sobre este tema en los distintos países.

(8) La Reunión consideró asimismo conveniente que se especifiquen los procedimientos aplicados a determinados productos que podrían dar lugar a

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problemas microbiológicos especiales por efecto del tratamiento con radiaciones y que se recomienden métodos de examen microbiológico para la investigación de estos problemas. En tales estudios podrían establecerse comparaciones con otros métodos de elaboración de productos alimenticios.

(9) La Reunión consideró asimismo que debe atenderse especialmente al problema de las alteraciones de calidad de los productos irradiados.

## RECOMENDACIONES

La Reunión consideró que el valor potencial de este nuevo método de conservación de alimentos justifica que se dediquen considerables esfuerzosa la investigación. Recomendó, pues:

(1) que los Estados europeos examinen atentamente los medios de fomentar y apoyar las investigaciones sobre irradiación de alimentos en todos sus aspectos;

(2) que a fin de mantenerse constantemente al corriente de la evolución que se registre en materia de irradiación de alimentos y de facilitar el desarrollo de la cooperación internacional en la investigación, establezca la FAO un grupo técnico permanente de trabajo sobre irradiación de alimentos, en cooperación, según proceda, con otras organizaciones internacionales interesadas, compuesto por expertos representantes de los Estados Miembros que se ocupen de la iniciación de programas de investigación y de actividades prácticas en el sector de la conservación de alimentos mediante radiaciones ionizantes o en su planeamiento;

(3) que en asociación con este grupo general de trabajo se establezcan los grupos técnicos que sean necesarios para atender a aspectos especiales de la irradiación de alimentos; uno de estos grupos técnicos podría crearse en el seno de la FAO y de la OMS para estudiar el problema de la formulación de principios fundamentales que regulen el empleo de la irradiación en la elaboración de alimentos y del desarrollo de métodos adecuados para el ensayo de alimentos irradiados a fin de determinar su inocuidad de modo que sea posible establecer una base común para la legislación sobre este tema en los distintos países. Podrían crearse otros grupos técnicos de esta naturaleza encargados de estudiar los aspectos microbiológicos y entomológicos de la irradiación de alimentos;

(4) que teniendo en cuenta lo costoso de las investigaciones y actividades prácticas en esta materia, se establezcan formas adecuadas de cooperación internacional en la investigación sobre irradiación de alimentos. En unión de otras organizaciones internacionales, la FAO debe encargarse de celebrar negociaciones, cuando así convenga, con Estados Miembros europeos y otros países interesados, para articular los oportunos sistemas de cooperación;

(5) que entre tales formas de cooperación internacional debe atenderse especialmente en estos momentos a la provisión de becas y de los medios adecuados en el plan internacional para la capacitación y las investigaciones en laboratorios experimentados en técnicas de irradiación de alimentos.

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# 1. OPENING ADDRESSES

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# Address of Welcome

B. F. J. SCHONLAND
Director, Atomic Energy Research Establishment,
Harwell

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LADIES and Gentlemen, on behalf of the Atomic Energy Authority, I should first of all like to welcome the delegates to this Conference at Harwell. We are very pleased that the Food and Agriculture Organization of the United Nations agreed to hold the Conference at Harwell because it will give the delegates an opportunity of seeing and commenting on our efforts in the general direction of food processing with radiation. Like all scientific workers our team in this field will greatly benefit from sharing ideas and hearing what is done elsewhere.

The United Kingdom Atomic Energy Authority has a mandate not only for developing nuclear power but also for exploring the possible applications of radioactivity in the research and industrial fields. In 1955 we set up the Technological Irradiation Group to investigate the uses to which powerful sources of radiation could be applied. We are at present in an experimental phase, not only because large sources are in short supply, but also because it is essential to pass through some years of experimental and development work before any large scale industrial projects can be contemplated. Even so, I am told there are possible large commercial demands for sterilization units which could use up half a million curies of $Co^{60}$, which is far more than we have immediately available. This may be optimistic; only time can show if they are feasible and economic.

The massive radiation sources are available in consequence of the Nuclear Power Programme and come to us in two ways. First by the production of $Co^{60}$ from the spare neutrons which are available in a power reactor during the running-in period, and secondly from the radioactive fission products obtained in the processing of nuclear fuel.

One section of our Technological Irradiation Group, which is situated at Wantage, is concerned with the radiation processing of food. In the programme of work we have been at pains to take advantage of the experience gained by the longer established bodies in the field of food technology. In particular, we have a very closely integrated programme with the Low Temperature Research Station at Cambridge. In the light of present developments it is interesting to recall the pre-war work of Dr. Lea at L.T.R.S. Until very recently the Cambridge Station was alone in Europe in the field with Dr. Hannan vigorously keeping the flag flying. I should like to acknowledge our indebtedness to that laboratory and also to the U.S.A. where the Quartermaster Corps has done so much and generously made the results of this important work available to us all.

This American work has been instrumental in stimulating the interest of many countries in radiation processing of food. Today we have I understand twenty countries represented here as well as the Food and Agriculture Organization itself, the International Atomic Energy Agency, the European Atomic Energy Commission and the Organization for European Economic Co-operation. There are also, I am told, about a dozen international scientific organizations represented.

Our own general programme has been laid down in consultation with a board of experts from the Universities and the Medical Research Council, who have been good

enough to put their experience at our disposal. You will have an opportunity of seeing the laboratories on one morning later in the week so I will not describe them in any detail. I shall only say that I understand that we have perhaps the most versatile collection of radiation sources that can be found anywhere. We have already begun to build a pilot plant with a capacity of 7 Mrad tons per day. This will be used in collaboration with industrial concerns for pilot feasibility trials. The materials to be processed will include medical equipment and general packaged goods including foods.

The subject of food processing by radiation has always been controversial. In Great Britain we have a reputation for making haste slowly and this is certainly going to be our policy in this matter. Before any processed foods can be offered to the public the Ministry of Health Expert Committee will have to be satisfied that no possible harmful effects can result and our animal farm is basically intended for feeding trials to meet this requirement. As you know, there are three general levels of dosage which can be applied to food: a low dose for insect control, a pasteurizing dose and a complete sterilizing dose. There are several promising applications for the lower dose treatments but it appears that many years of work must ensue before full sterilization can be achieved without making the food unattractive in the uncooked state. Forecasting is always dangerous but it is considered here that the low dose treatments which include the prevention of mould growth on fruit and the reproductive sterilization of insects in packaged goods will come to practical use many years before the full sterilization of food. It is likely that a partnership between radiation and some other treatment will be necessary for full sterilization.

You have I see a very full agenda so I will not take up any more of your precious time, and I shall close by wishing you an interesting and profitable week.

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# Opening Address

R. A. SILOW
Food and Agriculture Organization of the United Nations, Rome, Italy

On behalf of the Director-General of the Food and Agriculture Organization of the United Nations, I have pleasure in opening this European Meeting on the Use of Ionizing Radiations for Food Preservation.

First, Dr. Schonland, I should like to express, on behalf of the Director-General, and on behalf of all participants here present, our appreciation of your warm welcome to this country and to this mecca of all atomic scientists, your Atomic Energy Research Establishment.

This meeting is being convened in response to a recommendation of the First Meeting of the European Contact Group on the Uses of Isotopes and Radiation in Agricultural Research which was held at Wageningen in December 1956. That Contact Group was established by FAO, as part of the general United Nations programme for promoting the peaceful uses of atomic energy, in order to provide a medium for the exchange of information on national programmes and plans for the use of atomic energy in food and agriculture, with a view to encouraging European countries to make the fullest possible use of the important new atomic science techniques for the improvement of their agriculture and the raising of levels of nutrition of their peoples.

The Contact Group, at its Wageningen meeting, recognized that the use of ionizing radiations for food preservation was a highly promising field of application of atomic energy, but it also directed attention to the fact that many problems remain to be solved and much more information must be obtained before food irradiation can become a commercially practicable proposition in Europe. The necessary investigations would be difficult, lengthy and expensive in terms of money and the considerable number of trained research workers required in the many scientific and technical disciplines involved, and few countries in Europe would be in a position to undertake an adequate programme of research independently on the basis of their own resources. The Contact Group considered that advance was most likely to be achieved through some form of international co-operation in research of food irradiation and requested FAO to give consideration to specific means of developing such co-operation in Europe.

This present meeting is a first step in that direction. Its primary purpose at this stage is to survey the present status of food irradiation, and to attempt to evaluate the potential significance of the technique for European countries. Nothing is to be gained by an unrealistic over-enthusiasm that may lead to the belief that radiation can solve all problems of food preservation, and that there will no longer be any place for the conventional methods such as refrigeration, the use of heat and chemical preservatives, and canning. What this meeting seeks to do is to present an objective and factual review of present knowledge and accomplishments, on the basis of which European governments can draw their own conclusions as to the significance of food irradiation for their own particular conditions and requirements, and decide the extent to which they should become involved in work in this field.

It will be evident, therefore, that this meeting is not intended to be of the nature of a purely scientific conference, and the presentation of the experimental details of investigations would be inappropriate. The wide subject-matter field to be covered, and the limited time available, will make it

necessary for speakers to confine themselves to concise summary statements covering the general situation or indicating the general significance of work that has been done. The fact that many speakers have so kindly prepared written papers in which delegates can read fuller details later will do much to expedite discussion during the conference itself.

In preparing the provisional agenda FAO had the benefit of the advice of a small group of experts which met at FAO headquarters in Rome in April 1958. That expert group included:

(1) Dr. H. R. Barnell, Scientific Adviser's Division, Ministry of Agriculture, Fisheries and Food, Great Westminster House, Horseferry Road, London, S.W.1., England.
(2) Mr. S. Jefferson, Technological Irradiation Group, Atomic Energy Branch Research Establishment, Harwell, Didcot, Berks, England.
(3) Professor J. Kuprianoff, Federal Research Institute for Food Preservation, Karlsruhe, Kaiserstr. 12, Baden, Germany.
(4) Dr. D. A. A. Mossel, Central Institute for Nutrition Research, Catrynesingel 61, Utrecht, The Netherlands.
(5) Dr. Eirik Heen, Norwegian Fisheries Research Institute, P.O. Box 187, Bergen, Norway.
(6) Mons. P. Lévêque, Section for Applications of Radioelements, Department of Physiochemistry, Atomic Energy Commission, Saclay, France.
(7) Dr. G. T. Scarascia, National Nuclear Research Council, Via Belisario 15, Rome, Italy.
(8) Professor Aldo Mariani, National Institute for Nutrition, Rome, Italy.
(9) Dr. A. M. Paolucci, National Institute for Nutrition, Rome, Italy.
(10) Mr. B. Payne, International Atomic Energy Agency, Vienna, Austria.
(11) Dr. I. Nir-Grosfeld, Scientist in charge of Food Additives, Health Laboratory Methods Section, World Health Organization, Geneva, Switzerland.
(12) Mr. R. Thévenot, International Institute of Refrigeration, 177 Bd. Malesherbes, Paris, 17e, France.
(13) Mr. C. C. Thomson, The Organization for European Economic Co-operation, Paris, France.

In addition, since the first Provisional Agenda was distributed to governments in September, FAO has received many very helpful suggestions which have been incorporated as far as possible in a revised draft of that Provisional Agenda which will be submitted to delegates for approval. In considering that draft I will ask delegates to bear in mind that in preparing such a complex agenda various other arrangements equally satisfactory or better would undoubtedly have been possible, but that changes in the detailed time table that has been prepared would be extremely difficult at this stage. In asking delegates to be indulgent with regard to any short-comings in the agenda before them, for which short-comings FAO accepts full responsibility, I should also like to express our most profound gratitude to those many who have assisted us by submitting constructive and helpful suggestions.

After briefly reviewing the nature of the technique of food irradiation of the scientific principles on which it is based, a survey of the present status with regard to several of the main categories of food will be presented. In preparing for this meeting it has been recognized that whilst a considerable volume of basic research has already been undertaken in Europe, as will become evident during the proceedings, by far the greatest progress in investigations on the practical application of radiation to food preservation has been made in the United States of America. FAO is, therefore, grateful to have the services, as technical consultants, of three specialists from the United States: Dr. Herman F. Kraybill, Chief, Chemistry Division, U.S. Army Medical Research and Nutrition Laboratory, Denver, Colorado, U.S.A., Dr. B. S. Schweigert, American Meat Institute Foundation and Department of Biochemistry, The University of Chicago, Chicago, Illinois, U.S.A.; and Mr. George E. Tripp, Food Radiation Preservation

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Division, U.S. Food and Container Institute for the Armed Forces, Chicago, Illinois, U.S.A., who, with the kind co-operation of Dr. H. Lineweaver, Observer from the United States, will present accounts of present accomplishments in that country.

Brief statements on national programmes and plans in the United States and in European countries will then be followed by a discussion of the potentialities of food irradiation for European countries, on the basis of which consideration will be given to proposals for co-operation in research in Europe. Those proposals are the main objective of this meeting, and I would ask delegates to bear this objective in mind throughout the whole of the proceedings this week. Some of the possible forms of co-operation are outlined in the FAO report on "The Potential Contribution of Atomic Energy to Development in Agriculture and Related Industries" which is being distributed to delegates. The chapter on food irradiation in that report was distributed in mimeographed form to governments several weeks ago.

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It is realized that at this early stage delegates are not likely to be in a position to commit their governments to any course of action, but nevertheless some suggestions as to possible forms of co-operation can quite appropriately be put forward, and in fact it is to a representative international group of experts such as is present here that governments will look for guidance.

At that stage of the meeting consideration will also have to be given to future action that you may wish FAO to undertake in this field. In any future meeting on food irradiation in Europe it will not be necessary to go over the whole field in review again. In future FAO could probably make its most effective contribution by concentrating on specific limited areas of interest, and one of the objectives of the present meeting should be to identify such limited areas of interest in which international co-operation would be most rewarding. Examples of ways in which FAO could help would include the organization of technical working parties or committees on, say, microbiological problems of food irradiation; or biochemical aspects; or the possibilities of combination processes—that is, light radiation dosages in association with other methods of preservation; or the co-ordination of national legislation on the use of irradiated foods; or consideration of the experimental procedures on which such leglislation should be based, such as feeding tests, analytical methods, or dosimetry. These examples are only cited as possibilities, and you are asked to give thought, throughout this meeting, to the priorities which you wish FAO to attach to any form of help that we can give you in our future activities in this field.

At the last session of this meeting, next Friday afternoon, we hope to place before you, for your approval, a summary report of the proceedings of this meeting. My colleague, Mr. N. E. Holmes, Food Technologist in the Nutrition Division of FAO will act as Technical Secretary of this meeting and will be responsible for the preparation of that report. It is also hoped to publish the formal papers presented at this meeting in an appendix or supplement to that summary report.

A very interesting and stimulating programme lies ahead of us this week. In future years, this meeting may well come to be recognized as a most significant step, through international co-operation, towards the development of a new sector of the food industry in Europe, for it is the first intergovernmental meeting to be devoted exclusively to consideration of the potentialities of this new technique. It is appropriate that the first such meeting should be held in Europe, for this is the most compact grouping of countries, outside North America, potentially interested in the technique and likely to be technologically in a position to make use of it in the reasonably near future and, moreover, Europe is a region in which international co-operation is particularly needed. The large participation in this meeting is an indication of the extent of interest in food irradiation in Europe, and it is most gratifying to see that we have here almost one hundred delegates from 15 continental European countries, together with about 70 participants from the United

Kingdom, 6 observers from 5 non-European countries including the U.S.A., Australia, Pakistan, Canada and New Zealand, and 20 representatives of 14 interested international organizations. To all of you I extend a warm welcome and best wishes for a profitable meeting.

Amongst the international organizations present particular mention should be made of the International Atomic Energy Agency and the World Health Organization, sister agencies of FAO within the United Nations family, which also have special responsibilities on a world scale in connexion with the international programme for promoting the peaceful uses of atomic energy; and OEEC and EURATOM which have special responsibilities in this connexion in Europe. The representation of these and other international organizations at this meeting is especially welcomed because it will facilitate the co-ordination of the rapidly expanding international activities in atomic energy, which co-ordination is essential if financial and technical manpower resources are to be used efficiently and to full advantage.

In closing, Dr. Schonland, I should like to express, on behalf of FAO, our most sincere thanks to the Government of the United Kingdom, to the United Kingdom Atomic Energy Authority, and to you personally and those of the staff of your Establishment, both administrative and technical, who have been concerned with preparations for this meeting, for the generous facilities and cordial assistance that have been made available to us, and which we deeply appreciate.

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# Chairman's Address

NORMAN WRIGHT

Ministry of Agriculture, Fisheries and Food, 10 Whitehall Place, London, S.W.1.

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DR. Norman Wright, as chairman of the Conference, opened the technical part of the Symposium by welcoming all who attended on behalf of Her Majesty's Government, emphasizing that the United Kingdom felt it an honour to have been chosen as the venue for such a meeting. Dr. Wright regretted that he would be unable to attend the meeting in full, but he intended through his colleagues in the secretariat to be kept in close touch with the proceedings. In view of the fact that he would not be present all the time, he felt that it would be appropriate for him to propose the appointment of a vice-chairman, who would be available at all times to assist the sessional chairman and secretariat. (This was agreed at a suitable stage in the Agenda, when Dr. Kuprianoff was appointed vice-chairman.)

Dr. Wright went on to say that he felt it was very appropriate that this meeting should have been one of the FAO's first major activities in the atomic energy field; not only for the reason that it is a potentially important peaceful application of atomic energy, but because it is a field in which there would appear to be real opportunities of co-operation on an international basis. In this latter connexion, FAO had itself set an example in that the meeting was the result of co-operation of the FAO's Atomic Energy Branch with its own Nutrition Division. On a wider front the meeting indicated a very fruitful field for co-operation, not only between the various international agencies concerned, such as FAO, WHO, IAEA and OEEC but also between the individual countries of Europe who were represented at the Conference. In this connexion the chairman drew special attention to Items 7, 8 and 9 of the provisional Agenda which were as follows:

*Item* 7 Country Programmes and Plans

*Item* 8 Potentialities of Food Irradiation for European Countries

*Item* 9 Proposals for Co-operation for Research in Europe.

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# 2. TECHNOLOGICAL ASPECTS OF FOOD IRRADIATION

(a) Types, sources and applications of radiation
(b) Fundamental mechanism of radiation effects
(c) The effects of radiation on important constituents of food
(d) Microbiological aspects of food irradiation

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# Nuclear Sources of Radiation Energy

S. JEFFERSON
Technological Irradiation Group, Isotope Division,
Wantage Radiation Laboratory, Wantage, Berks

Radioactive cobalt is the most readily available source of $\gamma$-rays. 100–500 c are desirable for experimental work, but 50,000 c upwards are needed for treating bulk materials. The efficiency of utilization of the radiation depends upon the treated material occupying a maximum of the solid angle surrounding the source. Conveyors can be arranged to move packages around a source inside a concrete emplacement. Spent fuel elements form a useful source of $\gamma$-activity up to about 100 days from withdrawal from a high flux reactor. Radioactive caesium separated from fission products is another useful source of $\gamma$-rays having a half-life of about 27 years.

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THE practical considerations involved in radiation processing are the amounts of material to be treated, the range of dosage, and the accommodation of apparatus needed during the irradiation.

For experimental work on radiation effects a source of 100–500 c is desirable. Cobalt which has been irradiated in a reactor to become in part radioactive $Co^{60}$ is the most readily available type of source. The cobalt can be housed in a compact lead shield but, without extra shielding, this arrangement can only conveniently accommodate an irradiation space having about 500 $cm^3$ capacity.

It is generally more convenient to build a concrete shield of large enough dimensions to allow the experimental staff to enter the radiation chamber for the purpose of setting up apparatus associated with the irradiation. In such a chamber one wall can be made thick enough to act as a store for the radioactive material. As an alternative the radioactive material can be stored in a lead shield inside the irradiation chamber of a concrete shield. Arrangements must be made so that the radioactive source cannot be brought into the radiation chamber until a number of interlocks and safeguards have been brought into operation.

For treating bulk materials a source size of 50,000 c upwards of radioactive material is needed. The amount depends mainly upon three factors; the throughput of material being processed, the total dose needed for the process and the efficiency of utilization of the radiation. As an example, a source of 100,000 c of $Co^{60}$ can process 250 lb of material per hour with a dose of 2 Mrads, the efficiency being 40 per cent. Doubling the dose would naturally halve the throughput.

The time taken to absorb a dose of 1 Mrad in efficient arrangement of $Co^{60}$ is 5 to 10 hr. When food is being processed it is important that the inactivation dose for vegetative organisms (100,000 rads) should be given in a short enough time to prevent serious multiplication. The optimum temperature for multiplication is in the region of 37°C where the doubling time is about 20 min. At room temperature the rate is reduced to less than a quarter and so the doubling time becomes more than 1 hr. Although spores are more resistant to radiation, they are dormant and so the delay in acquiring the larger inactivation dose is not a problem.

In designing the process handling equipment around the radioactive source the maximum fraction of the solid angle round the source must be occupied. This is because the radiation leaves the source in all directions and any which reaches the shield

unimpeded is wasted. If the packages are small, not only should they occupy as much as possible of the solid angle around the source, but they should also be arranged in a second or even a third layer so that the radiation which penetrates the first layer without giving up all its energy is able partially to treat the next layer. Packages going through such an arrangement should traverse each layer in turn and also pass each side of the source so that the greatest uniformity of dose deposition is achieved.

The $Co^{60}$ so far used in the United Kingdom has been irradiated in the Windscale piles but it is planned in the future to introduce cobalt into the Central Electricity Generating Board reactors during the early part of their operating lives. During this time there are spare neutrons which would otherwise be absorbed in steel rods or wedges and so would not form a useful radioactive source.

The fission products formed during the burn-up of uranium fuel contain gamma-emitting radioisotopes and it is possible to use the fuel elements as a source of gamma rays 2 or 3 days after they have been withdrawn from a reactor. An experimental arrangement of this kind has been built at Harwell to make use of the spent fuel elements from the two high flux reactors DIDO and PLUTO. For industrial purposes the use of spent fuel elements is not very convenient because the average rate of decay of the fission products is rapid at first and so they have to be replenished at short intervals. In addition, to maintain a steady gamma flux in any one place it is necessary to rearrange the fuel elements as they decay. However, spent fuel elements do provide us with our first opportunity of having several millions of curies of gamma activity.

After the radioactivity of the spent fuel elements has decayed for about 100 days, they are then subjected to a chemical separation treatment primarily to recover the unused uranium. An extension of the plant can provide separated $Cs^{137}$ and $Sr^{90}$. $Cs^{137}$ is a useful source of gamma energy having a half-life of about 27 years. The gamma energy from $Cs^{137}$, however, is less penetrating than that of $Co^{60}$ and so is not a direct replacement for it; the lower energy also means that for a given dose rate more caesium is required. A plant is already under consideration for separating $Cs^{137}$ at the rate of about 1,500,000 c/annum commencing at the end of 1960.

The gaseous and volatile fission products from high temperature gas-cooled reactors can be used as sources of gamma radiation. There are plans for concentrating these fission products, which are mostly short-lived, on carbon beds. On account of the very short half-lives involved this form of source must be situated close to a reactor.

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# Machine Sources of Radiation

D. R. PERRY

Mullard Research Laboratories, Cross Oak Lane, Salfords, Redhill, Surrey

Electrons are shown to be the most suitable form of machine-produced radiation for food irradiation. The various methods of using electrons are compared and a summary of available methods of producing high energy electrons is given. Practical requirements, including radiation shielding, which would influence the choice of a machine are briefly discussed.

ELECTRONS are the form of machine-produced ionizing radiation most commonly used for irradiation. Heavy charged particles, such as protons, have to be accelerated to extremely high energies before they are sufficiently penetrating for most irradiation work and, in addition, such particles will cause considerable radioactivity in the irradiated material.

X-radiation can be produced by stopping accelerated electrons in a target. This radiation has a much greater useful range than the electrons which produce it and it is therefore sometimes used for the irradiation of thick articles. However, the production of X-rays is relatively inefficient and this technique is unlikely to be used in any economically attractive process.

The production of neutrons as a by-product of the primary radiation sets an upper limit of about 15 MeV for electron or X-radiation of food because of the risk of induced radioactivity.

## METHODS OF IRRADIATION USING ELECTRONS

Accelerating machines produce a thin beam of electrons which, for our purposes, must be allowed to leave the high vacuum inside the machine and pass out into the air, usually through a thin metal foil. A beam of electrons in passing through such a window and then through the air to the material to be irradiated is spread out by scattering with little loss in electron energy. This spreading out, alone, can be made to cover a sufficient area of material, but this method is normally too inefficient for economic use as 50 per cent or more of the electrons will be wasted. In order that the electrons shall be more evenly and therefore more efficiently spread over the material, it is convenient to deflect the beam rapidly to and fro so that it scans across the material carried on a conveyor moving at a speed dependent on the dose required. The scattering effect is useful for spreading out a scanning beam sufficiently to prevent the material from being irradiated in transverse strips separated by gaps which have received much lower doses than the remainder of the material.

Electrons with energies over 1 MeV have a maximum range of 0·5 cm/MeV in water, or more generally 0·5 g/cm²/MeV in any material of low mean atomic number. If the material irradiated has a thickness of about two-thirds of the maximum electron range, then about 80 per cent of the electron beam energy will be absorbed in the material. However, in order that the surfaces shall have a certain minimum dose, considerably higher doses must be given to the inside of the material and must be regarded as wasted energy. This reduces the effective efficiency of absorption to about 60 per cent which is near the best that can be achieved by single-sided irradiation.

If the material can be irradiated from both sides either simultaneously or in two separate runs then a considerable increase in efficiency may be obtained. If the thickness of material is $1\frac{2}{3}$ times the maximum electron range

(2½ times thicker than in optimum single-sided irradiation), an efficiency of about 85 per cent is obtained.

The efficiencies quoted above are approximately correct in the range of electron energies from 2 to 5 MeV. Somewhat higher efficiencies may be obtained if electron energies greater than 5 MeV are used.

The output of an accelerator is conveniently expressed in watts of electron beam power, obtained by multiplying the electron energy in megaelectron-volts by the mean beam current in microamps. The unit of absorbed radiation dose is the rad, which represents an absorption of 100 ergs/g of material. $10^6$ rads (or a megarad) therefore represents an absorption of $10^8$ ergs/g, 10 J/g or 10 W-sec/g. Therefore 1 kWh of electron beam energy fully absorbed in a material will impart a mean dose of 1 Mrad to 360 kg of material, or, in general:

$$1 \text{ kW} \equiv 360 \text{ kg Mrads/hr}$$

This relation is independent of electron energy, but must of course be corrected for the efficiency of energy utilization in order to calculate the quantities of material that may be irradiated with a given dose in a given time under given conditions.

It is interesting to compare a 1 kW electron accelerator with an isotope source capable of producing the same output, assuming equal efficiencies for both. It is easily shown that the equivalent isotope source is of the order of 100,000 c, whilst an accelerator with an energy of 30 kW is equivalent to approximately 3,000,000 c.

## TYPES OF MACHINE

(1) *Conventional high voltage power supplies* as used for X-ray sets are limited to about 500 kV. Commercially available equipment will produce free electron beams with energies as low as 50 keV, but the extremely short electron ranges (of the order of 1 mm even at 500 keV) are unsuitable for food irradiation except possibly for sprout inhibition on potatoes etc.

(2) *Cascade generators* use the Cockroft–Walton voltage multiplier circuit and apply the high potential obtained to an accelerator tube. A model with an electron energy of 3 MeV and an output of 15 kW has been suggested for commercial irradiation. The upper energy limit is set by insulation breakdown difficulties.

(3) *Resonant transformers* apply an alternating voltage to an accelerator tube and thus give a varying electron energy. Here also insulation problems set the upper energy limit, although a 4 MeV, 20 kW model is being developed.

(4) *Van de Graaff* machines are widely used for producing beams of charged particles, especially electrons, with energies between 1 and 3 MeV. In these machines electrical charge is conveyed by a rapidly moving insulating belt to a high voltage terminal from which electrons produced by a cathode are accelerated, in an evacuated tube, by the electrostatic field between the negative terminal and the other end of the tube which is at earth potential. A machine producing a beam power of 3 kW at 3 MeV is commercially available.

(5) *Linear electron accelerators*. Unlike all the previous types of machine these do not use very high voltages as the means of accelerating electrons and therefore are not limited, as far as maximum energy is concerned, by insulation breakdown difficulties. The electrons are accelerated by the electrical field due to a radiofrequency wave travelling along a special waveguide into which electrons are injected with energies of the order of 50 keV. In order that the accelerating field shall be high, pulsed sources of radiofrequency power are used. For example, a magnetron which gives a mean power of only 2 kW produces this power at the rate of 2 MW during a 2 $\mu$sec pulse.

The mean electron beam power in a linear accelerator can be up to 80 per cent of the mean r.f. power. In general, low efficiencies are obtained when high electron energies are produced with a given r.f. power source.

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The maximum electron beam power obtainable at a given energy may be increased either by using a larger single r.f. power source or by using a number of power sources injecting r.f. power at intervals along the accelerator.

Linear accelerators with beam powers up to 30 kW have been built or are under construction.

A very wide range of combinations of electron beam powers and energies have been obtained with or suggested for various types of machine, but for food irradiation the requirements are as follows:

(1) A selected energy up to 15 MeV
(2) High output
(3) High efficiency
(4) Convenience of installation
(5) Simplicity of operation and maintenance
(6) Low cost per kilowatt-hour of beam energy, which depends on the previous factors and also on the capital, running and maintenance costs.

## SHIELDING

It should be pointed out that, for a given output of irradiated material (expressed in kilogramme-megarads or similar units) much less shielding is required for an electron or beta-radiation source than for a gamma source.

The thickness of shielding required for a given type of source does not increase rapidly with the output of the source. For example, if the output of a 4 MeV accelerator is increased by a factor of 10 the thickness of concrete X-ray shielding walls around it will only have to be increased by 1 ft. This is one of the reasons for the development of high power machines which require less shielding per unit of output than smaller machines.

The quantity of shielding required is very dependent on how much equipment has to be within the shielded enclosure, and thus the physical size of an accelerator and its method of mounting are important considerations to be taken into account when comparing the economics of different types of machine and different arrangements of installation.

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# Gamma Radiation Dosimetry

D. B. DOVE

Technological Irradiation Group, Wantage Radiation Laboratory, Wantage, Berks

A discussion is given of the need for a simple reliable system of dosimetry, suitable for large scale use in the control of radiation processing units. The potentialities and limitations of a number of proposed systems are reviewed.

## INTRODUCTION

THE dosimetry of biological materials subject to gamma radiation of energy 0·2 to 2·0 MeV, is simplified by the fact that absorbed dose varies only slowly with material $Z$ number and photon energy. Energy is absorbed almost entirely by Compton interaction between incident photons and electrons in the medium, and hence energy absorbed per gram is proportional to the number of electrons per gram, i.e. to electron density. With hydrogen as an exception, materials of varying $Z$ number differ only slightly in electron density, and so difference in absorbed dose is largely due to relative hydrogen concentration. Thus for example, the electron densities of water, fat and bone are 3·34, 3·48, $3{\cdot}00 \times 10^{23}$, and the energies absorbed per gram are 93·2, 96·8, 83·5 ergs respectively, due to 1 r of 1 MeV radiation. In practice, this means that although dosimeters may have been calibrated against the energy absorption in one medium, only a small correction is required to obtain values of absorbed dose in other media.

Scattering of radiation at a point in a medium gives rise to energetic electrons which can dissipate energy at distances up to an electron range from the point. Hence discontinuities in a medium may give rise to effects at distances of the order of an electron range away; for a 1 MeV photon, the maximum Compton electron energy is 0·8 MeV and the corresponding range in water is ~0·4 cm. A dosimeter embedded in a medium may constitute a discontinuity, and electron equilibrium effects on the dosimeter should be considered, although it is only in rare cases that this is of importance.

## THE MEASUREMENT OF ABSORBED DOSE

The absorbed dose within a volume of material may be measured absolutely provided all absorbed energy appears as heat. A calorimeter constructed at the National Bureau of Standards, measures the energy locally absorbed in a graphite sphere, 1 cm in diameter, suspended inside a graphite shell. The temperature rise of the sphere is measured under adiabatic conditions and is related to absorbed energy by electrical calibration. More often the calorimeter is used to measure accurately the energy deposited within a volume of material inside the calorimeter, rather than the energy locally absorbed in a homogeneous medium. In this way, radiation induced changes may be calibrated against dose to provide further dose measuring devices. This is important since the calorimeter is unsuitable for the routine measurement of dose within a medium.

Various devices have been constructed which respond directly to dose rate, i.e. to energy absorbed per second. These include ion chambers, photovoltaic and conducting cells, and are not discussed here.

A notable effect that has been subjected to considerable study, is the oxidation of



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ferrous ions in 0·8 N sulphuric acid. This dosimeter requires care but yields results of good precision in the range $4 \times 10^3$ and $4 \times 10^4$ rads, and has become almost universally accepted as a secondary standard in this dose range (1 rad represents an energy absorption of 100 ergs/g, irrespective of absorbing medium or incident radiation). The upper limit of the dose range may be extended by bubbling oxygen through the solution before irradiation, or by adding copper ion. The ceric–cerous dosimeter covers the range $5 \times 10^4$ to $10^8$ rads but with possibly lower precision.

## PROCESS CONTROL DOSIMETERS

Neither of the above dosimeters is convenient for large scale use and a need exists for a simple reliable dosimeter suitable for routine control measurements in radiation processing. An effect suitable for dosimetry should be, within limits, independent of, or vary in a known manner with, dose rate, photon energy and environment, and be capable of reproducible measurement to the required degree of precision. Other properties, less essential but desirable in a process control dosimeter, are ease of measurement, low cost, chemical inertness, convenient size, ease of handling and good shelf life.

Within the dose range of interest in food treatment, viz. $10^4$ to $10^7$ rads, many systems of dosimetry have been proposed, which to a greater or lesser extent are suitable for process control, but none has yet received widespread acceptance. The following is a fairly comprehensive but not exhaustive compilation of reported systems. The dosimeters are grouped according to the effect being measured, and authors are to be found under the appropriate heading in the bibliography.

*Dosimeters based on optical density changes*

(1) *Glasses*

(a) A silver phosphate glass originally produced for use as a personnel dosimeter in the range 0 to $10^3$ rads, darkens considerably when subjected to higher doses. This radiation-induced darkening is proportional to dose up to $10^5$ rads; at higher doses sensitivity is reduced. The induced optical absorption is believed to be due to the formation of silver atoms within the glass due to silver ions acting as electron acceptors. There is a post irradiation recovery which necessitates correcting for exposure time, and time lapse before measurement, although to some extent this fading may be stabilized by post irradiation heat treatment (15 min at 100°C). Experiments in the low dose range indicate that energy response is uniform from 1·2 MeV down to 120 keV, rising by a factor of 18 at 65 keV. The darkening may be erased by heating to 400°C, enabling the glass to be re-used.

(b) Silicate glasses containing cobalt have been developed having good linearity of darkening up to $10^6$ rads with much improved stability. A flatter energy response is also expected since the effective $Z$ number is lower than that of the silver phosphate glass. Glasses containing other transition metals are reported to behave similarly.

(c) Antimonate glass shows no high dose saturation but actually increases in sensitivity above $10^7$ rads. Post irradiation heat treatment of 15 min at 125°C largely eliminates fading.

(d) The darkening of ordinary microscope slides has been used to map radiation fields.

(2) *Plastics*

(a) A commercial (Du Pont) blue dyed cellophane film has been found to decolorize irreversibly upon irradiation. An increase in transmission of approximately 1·5 per cent per Mrad is observed due to breakdown of the dye. The film is cheap, storage properties are good, and there is no post irradiation recovery. However, variations in film thickness (nominally

0·001 in.) and in dye concentration limit the accuracy attainable.

(b) Rigid non-plasticized polyvinyl chloride film develops a post irradiation coloration which is proportional to dose to at least $6 \times 10^6$ rads. Full development of the coloration requires heating for 4 hr at 120°F. The change is stable, film is cheap, available and easy to handle.

(c) Films of polyvinyl chloride having an incorporated indicator dye, methyl violet, have been prepared especially for dosimetry. The dye is radiation resistant but reacts with HCl liberated from the base material; dye retention vs. dose gives a straight line on a log-linear plot. These films cover the range $5 \times 10^5$ to $10^7$ rads.

(d) Experiments with polystyrene and polymethacrylate show that the ultraviolet absorption edge advances towards the visible region on irradiation. Increase in absorption measured at 4000 Å is linear with dose up to $2 \times 10^7$ rads. However, post irradiation bleaching is appreciable, being 50 per cent in 4 days for polystyrene.

(e) Attention at Wantage Radiation Laboratory has been concentrated on optical changes in commercially available materials, particularly on the radiation coloration reported to occur in I.C.I. Red 400, $\frac{1}{8}$ in. Perspex sheet. The radiation induced absorption is proportional to dose up to $2 \times 10^6$ rads. Post irradiation fading of the induced band occurs, but in adjacent parts of the spectrum a darkening takes place so that it is possible to choose the measuring wavelength, or band of wavelengths, in such a way that fading and darkening cancel. The value so obtained is stable over a number of days; there is, however, a long term recovery due to diffusion and thermal effects. Some post irradiation recovery is to be expected since it is believed that the dye is not decomposed, but acts as an electron acceptor, even though it is not initially ionized. Uniformity within a batch is satisfactory, but pre-irradiation storage conditions give rise to differences in response, which it is hoped may be reduced by thermal ageing, or other treatments, of new sheet.

Several dyes are known which decolorize irreversibly upon irradiation; these are, however, water soluble and are difficult to incorporate into a suitable base material. A solution to this problem might provide a dosimeter combining the handling properties of Perspex with the stability of the Du Pont cellophane.

(3) *Photographic film.* A special photographic print-out paper has been found which darkens with radiation up to $10^7$ rads. The darkening is stable, but response depends slightly on dose rate.

*Dosimeters based on viscosity changes*

(a) A "go–nogo" device has been produced which consists of a capsule containing styrene polyester and a metal ball. The polyester gels with a sharp end point, i.e. after a certain dose has been received. Movement of the metal ball after inverting the capsule shows whether or not gelation has occurred, and so indicates whether or not the end-point dose has been exceeded. About 30 min is required after irradiation for complete gelation.

(b) The decrease in viscosity due to chain cleavage in polymers and in polymers in organic solvents, has been measured against dose. Flow times are measured at a standard temperature in a viscometer, and change of flow time vs. dose is approximately straight on a linear-log plot. The dose range covered extends to $10^8$ rads.

*Dye solution dosimeters*

(a) A resorcinol stabilized chlorinated hydrocarbon-aqueous pH indicator dye system has been described for measurement of doses up to $10^6$ rads. Readings are obtained by titration.

(b) For high dose measurement dyes have been found which do not react with products of the solvent but which are decomposed

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directly by radiation. Such systems have reasonable sensitivity over the high dose range and are stable. Methylene blue in water has been used for some time, and more recently methyl yellow in cumene has been suggested. Although further work is required, such systems show promise for high dose measurement.

*Dosimeters based on luminescence*

The radiation degradation of an existing luminescence, and radiation induced luminescence, have been examined in various organic materials. The degradation of luminescence in anthracene occurs in the range 0·3 to $10^6$ to $200 \times 10^6$ rads and has been suggested for dosimetry.

*Dosimetry by gas decomposition*

The formation of nitrogen and oxygen from the radiation decomposition of nitrous oxide is proportional to dose up to $10^7$ rads, and nearly so to $10^8$ rads. The gas is thermally stable, and may be used, up to 200°C. Although unlikely to be of use for routine dosimetry, this method should prove of value in experimental gas phase dosimetry.

*Dosimetry by gas evolution*

Methods of utilizing gas evolution from aqueous solutions have been proposed. These may have specialized application but are not suitable for routine use.

## CONCLUSION

Although many effects have been suggested for dosimetry in the range $10^4$–$10^7$ rads, no single dosimeter has yet achieved universal acceptance. Several of these are unsuitable for large scale use, since the techniques involved are too complicated or time consuming, but may have other useful application.

The adoption of a system designed for dosimetry, such as the various glasses, will be influenced by eventual availability and cost. Several low cost commercial materials show useful radiation changes, e.g. Red 400 Perspex, Du Pont blue cellophane, but more work is needed before the full potentialities or limitations of these systems can be assessed.

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# Discussion

Dr. URI (U.K.): Would Mr. Perry indicate the maximum dose rate obtainable, for practical purposes, with the linear accelerator?

Mr. PERRY (U.K.): When discussing sources of pulsed radiation, such as linear accelerators, it is necessary to distinguish between the very high dose rate during a pulse and the mean dose rate measured over a period of time within which a large number of pulses have occurred. In the most intense part of the electron beam, *mean* dose rates of the order of 100 Mrads/sec may be obtained over a small volume of material. However, dose rate depends on the irradiation technique being used, and it is more convenient to think in terms of the quantity of material which can be treated in a given time. For example, a 1 kW machine operating with 60% irradiation efficiency has an output of 3·6 kg of material per minute at a dose of 1 Mrad.

Dr. ROBERTS (U.K.): In considering dose rate effects care must be taken in the case of machines giving pulses of electrons since effectively one is dealing with an intermittent radiation source. If the time interval between pulses is comparable with the lifetimes of any free radicals present in the system, then dose rate effects might conceivably be observed.

Mr. MURRAY (U.K.): Although 85 per cent of the available beam energy may be absorbed, with a satisfactory distribution of dose, by irradiating material from both sides, about 15 per cent of this is wasted in delivering a higher than necessary dose, and the theoretical maximum efficiency is of the order of 70 per cent.

In my experience there is little difference in the thickness of shield required for a 4 MeV, 2 kW machine and $10^5$ c of $Co^{60}$.

Dr. DE ZEEUW (Netherlands): As the path of electrons in tissue is relatively short, all the energy of the electrons is dissipated in a small volume, resulting in a rise in temperature. What order of temperature increase may be expected when using a linear accelerator?

Mr. PERRY (U.K.): The increase in temperature produced by radiation depends on the dose delivered and on the specific heat of the irradiated material, irrespective of electron energy or beam power of the accelerator. In the case of water, the temperature rise is 2·4°C/Mrad.

Dr. VOOGD (Netherlands): It is to be expected that the choice between the various sources of radiation for a process will depend largely on the special technical problems to be solved.

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# Effects of Radiations on Aqueous Systems under Aerobic and Anaerobic Conditions

J. WEISS
King's College, University of Durham, Newcastle-upon-Tyne

It is known that the chemical changes induced by ionizing radiations are, in general, considerably influenced by the presence of oxygen. It has been shown, that the presence of oxygen is not only capable of increasing the extent of radiation-induced oxidations but, in many cases, also leads to a *qualitative* difference in the nature of the radiation products. In particular, it has been found that in the presence of oxygen, more or less stable *hydroperoxides* are produced from various compounds, including many substances which occur in biological materials.

## 1. PHYSICAL AND CHEMICAL PRIMARY PROCESSES IN THE ACTION OF RADIATIONS ON WATER

CONSIDERABLE interest has centred around the chemical action of ionizing radiations (X-rays, $\gamma$-rays, $\alpha$-particles, neutrons, i.e. recoil protons) on aqueous systems; this, no doubt, has been due in part to the fact that biological systems are essentially aqueous in character. In 1944[1] it was suggested that the most important *chemical* primary process in the action of ionizing radiations on water is the formation of hydrogen atoms and hydroxyl radicals according to the *net* process:

$$H_2O \text{ -}\!\!\sim\!\!\rightarrow H + OH \qquad (1)$$

This simple equation has remained the key for our present understanding of the radiation chemistry of water.

In addition to the formation of H atoms and OH radicals (equation 1) it is now believed that there is some primary formation of molecular hydrogen and of hydrogen peroxide, the so-called "molecular yields".[2]

First, it may be of some interest to outline briefly certain suggestions relating to the physical primary processes leading to these chemical species.

It is well known that the energy loss in the absorption of ionizing radiations is due to excitation and ionization processes.[3] Excitation may lead to free radicals from excited water molecules according to:

$$H_2O^* \rightarrow H + OH \qquad (2)$$

With regard to the ionization processes, the formation of a positive ion according to:

$$H_2O^+ \rightarrow H_2O^+ + e \qquad (3)$$

and of negative ions by electron attachment:

$$H_2O^+ + e \rightarrow H_2O^- \qquad (4)$$

leading to what has been described elsewhere as a "hydrated electron" (somewhat similar to a "polaron" in an ionic crystal), are fairly obvious processes.[4] Questions arise, however, regarding the stability and possible reactions of these ions.[5] The life time of such ions with regard to dissociation into free radicals or atoms by interaction with the aqueous medium according to:

$$H_2O^+ \rightarrow OH + H^+ \qquad (5)$$

and

$$H_2O^- \rightarrow H + OH^- \qquad (6)$$

cannot be less than about $10^{-10}$ to $10^{-11}$ sec, i.e. of the order of the time of relaxation of water dipoles in liquid water. Furthermore, it is very likely that the relaxation time of the water molecules in the hydration shell of ions is considerably greater than in the pure liquid. It is possible, therefore, that these ions may have a life time long enough to enable them to undergo chemical reactions with the solutes, if only the concentration of the latter is sufficiently high and



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the relevant activation energies are sufficiently low.

It appears that certain features of the chemical primary processes in water can be explained by reactions of these ions.[6] For example, the increase in the primary radical yields, which has been observed in acid solutions, as compared with neutral solutions, may be due to a reaction between the "hydrated" electron and hydrogen ions:

$$H_2O^- + H_3O^+ \rightarrow 2H_2O + H \qquad (7)$$

thus tending to inhibit the primary recombination reaction according to:

$$H_2O^+ + H_2O^- \rightarrow 2H_2O \qquad (8)$$

A similar increase in the radical yield could also occur, in principle, at high pH owing to a reaction between $H_2O^+$ and hydroxyl ions.

The primary processes leading to the formation of the molecular products are not yet well understood. Some recent experiments[6] suggest that these may be formed by interaction of like ions according to:

$$2H_2O^+ \rightarrow H_2O_2 + 2H^+ \qquad (9)$$

and:

$$2H_2O^- \rightarrow H_2 + 2OH^- \qquad (10)$$

and/or by interaction of excited water molecules according to:

$$2H_2O^* \rightarrow H_2 + H_2O_2 \qquad (11)$$

## 2. THE ROLE OF MOLECULAR OXYGEN IN RADIATION-INDUCED REACTION

From the above considerations, it is now easy to see how oxygen may interact with the primary products, since oxygen can readily accept an electron, viz.

$$O_2 + H_2O^- \rightarrow O_2^- + H_2O \qquad (12)$$

and, in particular, can undergo a rapid association reaction with the hydrogen atoms produced by the radiation, viz.

$$H + O_2 \rightarrow HO_2\cdot \qquad (13)$$

In general, therefore, it is to be expected that oxygen will increase the yield of oxidation products. That oxygen can enhance oxidation processes without qualitatively changing the reaction products can be seen very clearly in some simple inorganic systems, as, for instance, in the radiation-induced oxidation of ferrous to ferric ions, which has been studied in detail.[7]

In the case of organic solutes, the influence of oxygen on the oxidation yield may be due to a number of factors. The initial reaction of OH radicals with a solute (RH) can, in many cases, be represented as a dehydrogenation step of the form:

$$RH + OH \rightarrow R\cdot + H_2O \qquad (14)$$

In the absence of dissolved oxygen, the organic free radicals (R·) could then react according to: (X· e.g. an H atom):

$$R\cdot + X\cdot \rightarrow RX \quad \text{(recombination)} \qquad (15)$$

$$2R\cdot \rightarrow R_2 \quad \text{(dimerization)} \qquad (16)$$

$$2R\cdot \rightarrow RH + R' \text{ (dismutation)} \qquad (17)$$

The presence of molecular oxygen will, in general, influence the course of the radiolysis because of (i) its reaction with hydrogen atoms to form $HO_2$ radicals according to equation (13) and (ii) interaction with the organic radicals according to:

$$R\cdot + O_2 \rightarrow RO_2\cdot \qquad (18)$$

$$R\cdot + O_2 \rightarrow R' + HO_2\cdot \qquad (19)$$

In this manner reactions (15) to (17) can be suppressed.

The inhibition of dimer formation by molecular oxygen, for example, has been clearly demonstrated in the irradiation of aqueous ethanol solutions.[8] Here, in the absence of oxygen, 2:3-butanediol is produced by the reaction sequence:

$$CH_3\cdot CH_2OH + OH \rightarrow CH_3\cdot CHOH + H_2O \qquad (20)$$

$$2CH_3\cdot CHOH \rightarrow CH_3\cdot CH(OH)\cdot CH(OH)\cdot CH_3 \qquad (21)$$

Formation of this diol is completely suppressed in the presence of oxygen, evidently owing to the production of the peroxy radical ($CH_3 \cdot \underset{O_2}{\dot{C}}HOH$) which, as we have found in this particular case, will lead to acetaldehyde and hydrogen peroxide.[8]

It may be of interest to note, as has been pointed out previously,[1] that there is no fundamental difference between the direct and indirect effects of the radiation. An R· radical can be produced, according to equation (14) by interaction between RH and an OH radical, but also by the direct effect of the radiation on the RH according to the chemical primary process:

$$RH \longrightarrow R\cdot + H \qquad (22)$$

The R· radical produced in this latter process will again tend to combine with molecular oxygen to form an $RO_2$ radical (equation 18). It can, therefore, be stated that the differences between direct and indirect effects are, in general, only of a quantitative nature.

In several aqueous systems of organic solutes it has been observed that the radiation yield of a particular oxidation product is between *three* to *six* times as great in oxygen-containing solutions, as in the absence of oxygen, the actual values depending on the quality of the radiation and on the solute.

Concerning the role of the $HO_2$ radicals, they are less "oxidizing" than the OH radicals and appear to be relatively unreactive, particularly towards saturated organic substances. This is, in fact, to be expected on thermodynamic grounds, since the bond dissociation energy of water, D(H–OH) = 118 kcal, whereas the bond dissociation energy of hydrogen peroxide, $D(H\text{–}O_2H)$ = 90 kcal. Moreover, particularly in neutral solutions, the hydroperoxy radical may act as a reducing agent in the form of its anion from the dissociation equilibrium:

$$HO_2 \rightleftharpoons H^+ + O_2^- \qquad (23)$$

In most cases, the fate of the $HO_2$ radicals is a reaction with an organic peroxy radical,

e.g. $CH_3 \cdot \underset{O_2\cdot}{\dot{C}}HOH + HO_2 \rightarrow$

$$H_2O_2 + O_2 + CH_3 \cdot CHO \qquad (24)$$

and/or dismutation to form hydrogen peroxide, viz.,

$$2HO_2 \rightarrow H_2O_2 + O_2 \qquad (25)$$

Although, under certain conditions, $HO_2$ radicals appear to be able to add to an ethylenic double bond.[9]

In the course of our studies, we have encountered several cases where the presence of oxygen during irradiation led not only to the expected *quantitative* differences in the efficiency of the oxidative process but also to some *qualitative* differences in the reaction products.

For instance, the irradiation of oxygenated aqueous solutions of benzene yielded mucondialdehyde[10,11] and the irradiation of tryptophan solutions in the presence of oxygen gave formylkynurenine.[12] Neither of these products was found under anaerobic conditions.

Other notable examples are the radiation-induced conversion of ethyl phosphate into acetyl phosphate[13] and the formation of formaldehyde from the α- and β-glycerophosphates, which occurs only in the presence of oxygen.[14]

It has been concluded that the important reaction step which leads to these specific products is again the formation of organic peroxy radicals. The subsequent reactions of the organic peroxy radicals in these aqueous systems are not known with certainty. Since the formation of peroxides and hydroperoxides is known to occur in the autoxidation of various inorganic and organic compounds, it seemed highly probable that similar intermediates would occur in the chemical action of ionizing radiations in the presence of oxygen. We have previously suggested that the formation of mucondialdehyde from benzene and also of formylkynurenine from tryptophan may be the result of the decomposition of unstable hydroperoxides formed via the organic peroxy radicals. Recent detailed studies (G. Scholes, J. Weiss and C. M. Wheeler, unpublished

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data and ref. (15)) on the radiolysis of ethyl phosphate solutions have indicated that the mechanism of formation of the acyl compound involves intermediary organic alkoxy radicals (RO·) which can be formed from the corresponding $RO_2$ radicals:

$$CH_3{\cdot}CH_2{\cdot}OP(OH)_2 + OH \rightarrow CH_3{\cdot}\dot{C}H{\cdot}OP(OH)_2 + H_2O \quad (26)$$

$$CH_3{\cdot}\dot{C}H{\cdot}OP(OH)_2 + O_2 \rightarrow CH_3{\cdot}CH{\cdot}OP(OH)_2 \quad (27)$$
$$\dot{O}_2{\cdot}$$

$$2CH_3{\cdot}\dot{C}H{\cdot}OP(OH)_2 \rightarrow \quad (28)$$
$$\dot{O}_2{\cdot}$$

$$2CH_3{\cdot}CH{\cdot}OP(OH)_2 + O_2$$
$$\dot{O}{\cdot}$$

$$CH_3{\cdot}\dot{C}HOP(OH)_2 + O_2 \rightarrow$$
$$\dot{O}{\cdot}$$

$$CH_3{\cdot}COP(OH)_2 + HO_2{\cdot} \quad (29)$$
$$\ddot{O}$$

acetyl phosphate

Hence, it is not unlikely that in the case of the radiation-induced oxidation of benzene and of tryptophan the corresponding alkoxy radicals may be the intermediates responsible for the products found in oxygenated solutions.

## 3. THE RADIATION-INDUCED FORMATION OF ORGANIC HYDROPEROXIDES

In view of the possible importance of organic hydroperoxides and peroxides in the radiolytic oxidation of organic compounds as well as in radiobiological processes, it was decided to investigate in some detail the formation of these compounds under the influence of ionizing radiations. Attempts were made to find systems where the hydroperoxide would be relatively stable and could be identified as such.

The first case where this was posssible was with aqueous solutions of methane in which the radiation-induced formation of methyl hydroperoxide was established.[16] It was suggested that this was formed according to equations (30) to (32).

$$CH_4 + OH \rightarrow CH_3{\cdot} + H_2O \quad (30)$$

$$CH_3{\cdot} + O_2 \rightarrow CH_3O_2{\cdot} \quad (31)$$

$$CH_3O_2{\cdot} \xrightarrow{\text{reduction}} CH_3O_2H \quad (32)$$

There are various possibilities for the formation of methyl hydroperoxide from the $CH_3O_2{\cdot}$ radical (equation 32); in view of the observed pH-dependence we have suggested that this reduction process is brought about by the anion of the $HO_2$ radical, as in equation 33.

$$CH_3O_2{\cdot} + O_2^- \rightarrow CH_3O_2^- + O_2 \quad (33)$$

$$CH_3O_2^- + H^+ \rightleftarrows CH_3O_2H \quad (34)$$

Subsequently, hydroperoxide and peroxide formation have been studied in a number of different systems; a brief account is given here of some of the results obtained.

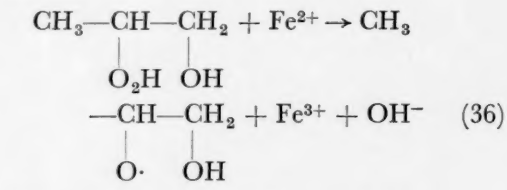
### A. *Unsaturated hydrocarbons*

The radiolysis of aqueous solutions of ethylene/oxygen and of acetylene/oxygen mixtures yields only very small amounts of stable hydroperoxides.[9] With propylene as a solute, on the other hand, very good yields of a hydroperoxide can be obtained up to $G \sim 2{\cdot}6$ (P. G. Clay, J. Weiss and J. Whiston, unpublished data). One may presume that here a hydroxy-hydroperoxide is formed according to the reaction sequence:

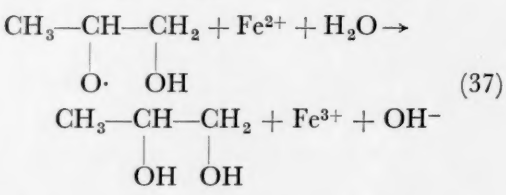
$$CH_3{-}CH{=}CH_2 \xrightarrow{OH} CH_3{-}\dot{C}H{-}CH_2{-}OH \xrightarrow{O_2} CH_3{-}CH(O_2{\cdot}){-}CH_2{-}OH \xrightarrow{\text{reduction}} CH_3{-}CH(O_2H){-}CH_2{-}OH \quad (35)$$

Some experimental support for this particular scheme follows from the observation that, on treatment of the irradiated solution with an excess of ferrous salt, two ferric ions are produced per hydroperoxide group decomposed, and propylene glycol is formed. This

process can be represented by equations 36 and 37.



(36)



(37)

B. *Nucleic acids and related compounds*

On irradiation of aqueous solutions of yeast ribonucleic acid (RNA) and of thymus deoxyribonucleic acid (DNA) in the presence of oxygen it has been found[18] that more or less stable hydroperoxides are formed; these are produced in amounts which are quite significant, i.e. with a yield of $G \sim 0{\cdot}5$ ($G$ = molecules/100 eV). In DNA solutions, the post-irradiation decay of the hydroperoxide can be described by a superposition of two first-order processes, suggesting that there are two different hydroperoxides present which decay at different rates.[17] Subsequent investigations of a number of ribonucleotides and of the corresponding purine and pyrimidine bases have shown that the formation of hydroperoxides is associated with the pyrimidine bases.[18]

Thymine, in particular, has been found to give a hydroperoxide that is comparatively stable in neutral aqueous media. The thymine hydroperoxide can be separated and isolated from the solution by ion-exchange chromatography. It has been found that in the hydroperoxide the characteristic ultraviolet absorption associated with the 4,5 double bond of thymine is no longer present. Mild hydrolytic breakdown of the hydroperoxide yields a compound which gives a dihydrazone with 2:4 dinitrophenylhydrazine. This, in conjunction with the studies reported earlier[19] would suggest that, in the radiation-produced hydroperoxide, the 4,5 double bond has been saturated by the addition of *one* OH and *one* $O_2H$ group, respectively. The radiation-induced production of hydroperoxides in the pyrimidine bases of nucleic acids may be of considerable significance, since, if in fact, the formation of hydroperoxides does occur on the genetic material *in situ*, this is bound to have far-reaching biological consequences.

C. *Amino acids, peptides and proteins*

In a recent investigation of a series of amino acids and simple peptides[20] evidence has been found, in many instances, for the radiation-induced formation of relatively stable hydroperoxides. The amino acids studied appeared to fall into three groups. The highest yields of organic hydroperoxide were obtained from solutions of some naturally occurring amino acids containing a tertiary carbon atom, e.g. *iso*leucine, or valine. Lower yields were obtained from amino acids which contain not less than two adjacent $-CH_2-$ groups (glutamic acid, arginine) whereas some of the simple amino acids (e.g. glycine, alanine) and those containing a benzene ring (e.g. phenylalanine), as well as histidine and hydroxyproline, did not yield any appreciable quantities of a hydroperoxide under the prevailing experimental conditions ($10^{-3}$ to $10^{-2}$ M solutions, irradiated with X-rays at pH 4 to 6).

Of various di- and tri-peptides examined, only those containing a leucyl residue yielded an organic hydroperoxide. It is suggested, therefore, that hydroperoxide groups may be formed preferentially in proteins that contain amino acid residues which themselves are known to form hydroperoxides. This view has been confirmed by some recent experiments with aqueous solutions of trypsin (G. Scholes, J. Ward and J. Weiss, unpublished data) which give a hydroperoxide yield of $G \sim 0{\cdot}6$.

D. *Steroids*

A number of steroids have also been investigated, including cortisone, cortisol, prednisolone, prednisone and deoxycorticosterone (DOC). All these substances yield hydroperoxides on irradiation of their aqueous

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solutions with X-rays in the presence of oxygen (R. E. OAKEY and J. WEISS, unpublished data).

Particular attention has been paid to deoxycorticosterone, which gives a relatively high yield of an organic hydroperoxide, of $G \sim 2$ at pH 7. Post-irradiation decay studies indicate the presence of two distinct peroxidic functions, formed in the ratio of approximately 2 : 1. The more abundant hydroperoxide appears to be rather more stable in neutral solution (half-life ~17 hr) than the minor constituent (half-life ~0·5 hr). In order to elucidate further the mechanism of the formation of these hydroperoxides in these steroids, irradiations have been carried out in the presence of molecular oxygen labelled with the $O^{18}$ isotope. Isotopic analysis of the radiation products has confirmed that $O^{18}$ labelled oxygen enters into the molecule. It is reasonable to suppose that the $C^{IV}$–$C^{V}$ ethylenic bond and also the side chain are rather favourable sites for initial attack by OH radicals; this is supported by the observation that the concomitant chemical changes include (i) the elimination of the ultraviolet absorption associated with the chromophoric ($\alpha,\beta$-unsaturated ketone) grouping in ring *A*, and (ii) disappearance of the characteristic reaction of the side chain with tetrazolium blue. It has been found that there is a quantitative correspondence between the yield of the more stable hydroperoxide and the extent of the destruction of the chromophoric grouping and, between the yield of the less stable hydroperoxide and the disappearance of the side-chain.

## CONCLUSION

The chemical effects of ionizing radiations on aqueous systems can be considerably influenced by the presence of molecular oxygen. Oxygen can, in general, not only increase the extent of radiation-induced oxidation but, as has now been shown in many cases, may also lead to a *qualitative* difference in the nature of the radiation products. It is suggested that this latter effect is mainly due to the intermediate formation of organic peroxy radicals ($RO_2\cdot$) which are formed by the association of organic radicals (R) with molecular oxygen ($R + O_2 \rightarrow RO_2$).

In particular, it has been found that, in the presence of molecular oxygen, more or less stable organic hydroperoxides can be formed from various solutes, including many substrates of biological interest. Among these are:

(i) *nucleic acids* (DNA and RNA) and related compounds, e.g. pyrimidine bases such as thymine and uracil;

(ii) *amino-acids* (e.g. leucine, valine, glutamic acid, arginine); *simple peptides* (glycyl-leucine,leucyl-glycyl-glycine); *proteins* (e.g. trypsin);

(iii) *steroids* (e.g. deoxycorticosterone, cortisol, cortisone).

In many instances, evidence relating to the chemical structure of the radiation-produced organic hydroperoxides has been obtained. For example, in the case of the thymine hydroperoxide it is suggested that it is a hydroxy-hydroperoxide, formed by saturation of the 4,5 double bond.

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# Some Aspects of Radiation-induced Free Radical Reactions and the Significance of Free Radical Acceptors

N. URI

Ministry of Agriculture, Fisheries and Food, Scientific Adviser's Division (Food), Research Establishment, Greyhope Road, Aberdeen

The exact nature of the radiation–chemical primary act is controversial but there is little doubt that a major role is played by secondary reactions in which free radicals participate. Some of the most undesirable side effects in food irradiation would be produced by free radicals, particularly when they can cause a chain reaction. The study of radiation-induced oxidative rancidity, which is a chain reaction, and its inhibition by free radical acceptors is, therefore, both of fundamental and of practical importance. In order to reach the backbone of the mechanism it is advantageous to work with pure linoleic acid and its esters. Some protection against radiation effects can be achieved by energy transfer, but this mode of action could not be efficient once free radicals are formed. A detailed study of free radical acceptors is, therefore, necessary. Some of their essential properties are outlined and a comparison is made between their efficiency in thermal oxidative rancidity and radiation-induced oxidative rancidity. This outline includes newly synthesized polyhydroxyflavones and special emphasis is placed on the apparent significance of amino derivatives. The importance of dose rate is also indicated and application to milk is mentioned. Finally, the concept of "radio-antibiotics" is introduced and the prospects of efficient free radical acceptors are summarized.

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WHEN I compiled a critical review on inorganic free radicals in solution seven years ago*, I included a brief chapter on radiation chemistry, in which I described the subject as relatively new. A good deal of research has been carried out in many centres since then, but I still hold the belief that we are nowhere near to recognizing the underlying mechanisms as precisely as in thermal or photochemical reactions. The modern tendency of the kineticist is to be dissatisfied with his insight until he can break up a set of reactions into single steps, generally involving bimolecular collisions. This has been achieved with a fair degree of success in many systems including photochemical reactions, but in radiation chemistry one still has, very frequently, to be content with the recognition of overall phenomena while the individual steps remain a subject for speculation and controversy.

I consider it a privilege to follow Professor WEISS in this series of lectures, because he has been responsible for so much pioneering work in the whole field of free radicals. The paper published by HABER and WEISS[1] in the *Proceedings of the Royal Society* in 1934 has been the foundation of many investigations in the fields of reaction kinetics, photochemistry, electrochemistry and radiation chemistry. In this paper HABER and WEISS made a kinetic study of the reaction of ferrous ion and ferric ion with hydrogen peroxide and found it possible to interpret the kinetic data in terms of free radical mechanisms involving OH and $HO_2$ radicals. These concepts were introduced by WEISS into radiation chemistry in 1944.[2] This

* *Chem. Rev.* **50,** 375 (1952).

is at least the first published account which gave rise to a new development in radiation chemistry. The assumption was made that, whether the primary species were ions or excited molecules, it is free radicals which are ultimately responsible for the secondary reactions. $H_2O^+$ ion would split into $H^+$ and $OH^\bullet$ and, similarly, $H_2O^-$ would lead to the production of H atoms.

One of the main difficulties, however, of translating the concepts gained in the field of free radical chemistry to ionizing radiations is to be found in the fundamental radiation–chemical phenomenon of the lack of specificity. Since energy from ionizing radiations is indiscriminately absorbed, in sharp contrast to the properties of light of certain wavelengths, in many systems the multiplicity of radicals present makes an accurate kinetic study very difficult indeed. Radiation induced free radical reactions are, therefore, much more difficult to handle even for relatively experienced researchers and very frequently the reward is no more than some new qualitative insight, in the place of a harmonious correlation of facts producing a quantitative picture. Perhaps it is this multiplicity of reactions which makes it more difficult to obtain strictly reproducible results and even more so, agreement between various laboratories. Apart from such fundamental disagreements as, for example, the yield of the ferrous sulphate dosimeter, which lasted for years before being resolved, quantitative agreement is frequently lacking in basic aspects, e.g. the very important problem relating to the uniformity of free radical distribution; this was recently pinpointed by DAINTON[3] referring to a symposium on radiation chemistry held in Paris a few years ago. In the system solvent–diphenyl picryl hydrazyl (DPPH) the uniformity of the solvent radical distribution affects the competition between the DPPH–solvent radical reaction and the solvent radical recombination. In a detailed study of such a system carried out by WILD[4], only in one out of ten solvents was fair agreement obtained with Chapiro. As far as we are concerned we cannot reproduce results obtained by MEAD in California[5,6] for the irradiation of emulsions of methyl linoleate; the doses with which he claims to have obtained significant radiation effects are, in our experience, so small that results are difficult to distinguish from non-irradiated blanks.* On the other hand, we obtain tolerable disagreement, if not fair agreement in the irradiation of pure methyl linoleate with the results reported by LUNDBERG *et al.*[7]

Before discussing this subject in great detail, I shall first try to explain my basic approach, which must be shared by many who work with a practical aim in mind. In the preceding paper we heard Professor WEISS outlining his academic research, which is, in the main, and rightly so, fundamental research for the sake of knowledge. However, there is a second type of fundamental research, namely one in which the problems of study are so selected that they are most likely to give beneficial results, which can be applied. Perhaps too little of this type of research is being pursued, in practice, although its importance is fully recognized. The contents of my paper comprise a general review, together with some of our own results and, therefore, I am bound to explain our main approach.

We have been concerned, during the last few years, with the problem of oxidative rancidity and its inhibition. It is well recognized that oxidative rancidity is due to a chain reaction, involving free radical intermediates, of the type

$$\left.\begin{aligned} \rightarrow\ & R^\bullet + O_2 \rightarrow RO^\bullet_2 \\ & RO^\bullet_2 + RH \rightarrow ROOH + R^\bullet \end{aligned}\right]$$

One of our main subjects of research has been the continuous improvement of existing free radical acceptors which inhibit oxidative rancidity and the production of new and more powerful ones. If, indeed, the undesirable radiation effects, such as off-flavours and off-odours, are produced by free radicals

* Note added in proof: Further investigation is required to establish whether the discrepancies are not entirely due to the dose rate effect described in this paper.

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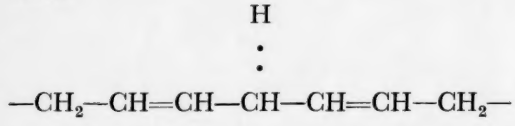
there is, quite clearly, a common objective in the study of oxidative rancidity in general, radiation induced oxidative rancidity and food irradiation in general. Many food components are, of course, liable to be affected by radiation, but it is obvious that in those cases where free radicals can give rise to chain reactions, the effect will be very marked. Even if chain reactions were not the most harmful ones, they can be more readily investigated because a relatively small concentration of free radicals gives rise to the production of a substantial quantity of the final products. In general it is linoleic acid with its 1:4-diene structure and the resulting weak α-methylenic C . . . H bond



which is responsible for the oxidative rancidity of fats. It appears that in order to study the efficiency of free radical acceptors in the radiation-induced secondary free radical reactions, linoleic acid and its esters would be very suitable substrates, particularly under normal aerobic conditions. Polymerization and degradation of polymers are alternative systems, which have in fact been investigated, but it is not unlikely that the study of the aerobic oxidation of linoleic acid and its esters, e.g. methyl linoleate, has a more direct bearing on the problems encountered in food irradiation. Indeed, radiation-induced oxidative rancidity is bound to be a major obstacle to the successful application of the radiation sterilization of food. This applies not only to the formation of hydroperoxides and their carbonyl decomposition products but also to the fact that the free radical intermediates continuously produced in the course of the autoxidation, give rise to the destruction of most pigments and vitamins. If one were to find a very efficient non-toxic protector against the radiation-induced aerobic oxidation of methyl linoleate, I would be surprised if this were not an important major development in the whole field.

I should like to emphasize that I have not overlooked the other important mode of protection, namely energy transfer. This is an accepted concept in photo chemistry, well illustrated by BOWEN's example[8] of mixtures of aromatic hydrocarbons where traces of naphthacene may, by their presence completely suppress and replace the photo-excited fluorescence of anthracene (the former being green and the latter being blue). In photosynthesis, it is now considered that energy absorbed by β-carotene is transferred to chlorophyll *a*. If such energy transfer is possible in radiation chemistry one readily visualizes protection by the replacement of efficient or harmful radiolysis of one compound by a less efficient or harmless radiolysis of another. In fact, this was convincingly demonstrated with the use of deuterium tracer in *cyclo*hexane–benzene mixtures and also in *cyclo*hexane solutions of iodine and iodides by BURTON *et al.*[9] It is assumed that it is the "molecular" yield of $H_2$ which is mainly affected, but the "free radical" yield, i.e. hydrogen production via free radical intermediates, is less affected. One is, therefore, bound to think again in terms of producing efficient free radical acceptors. In our own work, therefore, we concentrate on examining the efficiency of free radical acceptors in inhibiting both peroxide and carbonyl formation during the radiation induced aerobic oxidation of linoleic acid.

Having obtained some remarkable results in the inhibition of the thermal aerobic oxidation of linoleic acid, an attempt has been made to apply this experience to radiation-induced aerobic oxidation. It is generally accepted that the inhibition reaction in the autoxidation is due to a reaction of the $RO^{\bullet}_2$ radical with the free radical acceptor

$$RO^{\bullet}_2 + AH \rightarrow ROOH + A^{\bullet}$$

The $A^{\bullet}$ radical is usually of the semiquinone

type and is inefficient in propagating the chain reaction and ultimately would disappear in a disproportionation reaction reproducing in part the original antioxidant. There are many factors which determine the efficiency of the antioxidant or free radical acceptor (these are discussed in a forthcoming publication[10]) but foremost amongst them are: (1) the activation energy of the above reaction which is dependent on the bond dissociation energy of A...H and is also linked with the oxidation–reduction potential; (2) steric factors for the particular reaction; (3) stability of the A• radical, and (4) the partial disappearance of the antioxidant AH by the "forbidden route" of a chain reaction. Bearing in mind all these factors and also bearing in mind the desire to keep to structures closely related to naturally occurring compounds, we found wide scope in exploring the free radical acceptor efficiency of hydroxyflavones and some of their alkyl derivatives. The antioxidant activity of these was previously recognized[11] but never fully explored in systematic detail and in some ways any previous efforts were directed towards the wrong derivatives, because the relative importance of chelating and free radical acceptor properties had been misunderstood. The flavones which were found to be most efficient in our investigations were the 3:7:8:2′:5′- and 3:5:8:3′:4′ -pentahydroxyflavones.[12,13] These showed antioxidant efficiencies up to fifty times that of propyl gallate in the aerobic oxidation of linoleic acid. It is believed that 3:5:8:2′:5′ -pentahydroxyflavone has been synthesized (by Dr. Thomas) and its activity is now being examined in detail.

Though protective effects were recorded it must be said that, unfortunately, these remarkable factors in relation to other antioxidants could not be repeated in an analogous manner in the radiation-induced aerobic oxidation of linoleic acid and methyl linoleate. Further research is necessary to give the answer to the obvious question: why?

Our first radiation experiments were carried out with a $Co^{60}$ hot-spot source which we obtained on loan from Harwell and for which I should like to express my appreciation, particularly to Mr. Horne, who negotiated the loan. Recently we obtained our own source of 590 c, the shielding of which is based on a Harwell design, which has been modified and improved in Aberdeen.

In Table 1 I present results which were obtained during the last few months, in the study of the effect of free radical acceptors on the aerobic oxidation of methyl linoleate. I must say that I believe that these results are not yet sufficiently numerous to give a clear picture and have not yet

TABLE 1. Irradiation of methyl linoleate in the presence of air*
(Dose rate = $4 \times 10^5$ rads/hr)

| Antioxidant (Usual conc. = M/1000) | $10^5$ rads Peroxide/Carbonyl (mM/kg) | | $5 \times 10^5$ rads Peroxide/Carbonyl (mM/kg) | | $2 \times 10^6$ rads Peroxide/Carbonyl (mM/kg) | |
|---|---|---|---|---|---|---|
| None | 2·38 | 0·08 | 8·56 | 0·41 | 34·1 | 3·91 |
| Propyl gallate | 0·70 | 0·59 | 2·87 | 0·61 | 14·2 | 3·22 |
| 6-ethyl,3:7:2′:5′-tetrahydroxyflavone | 1·54 | 0·59 | 3·79 | 0·70 | 14·2 | 3·02 |
| BHA | 0·94 | 0·51 | 2·72 | 0·86 | 11·1 | 3·40 |
| Carboxymethylmercaptosuccinic acid | 3·00 | 0·21 | 10·4 | 0·99 | | |
| Diphenyl *p*-phenylenediamine | 0·44 | 0·17 | 1·88 | 0·58? | 5·56 | 1·14? |
| | 0·43 | | 1·88 | | 7·20 | |
| 7:8-dimethoxy,3:2′:5′-trihydroxyflavone | 1·50 | 0·18? | 3·73 | 0·66? | 12·9 | 1·48? |
| 3:7:8:2′:5′-pentahydroxyflavone | 0·82 | 0·61 | 2·98 | 1·47? | 9·60 | 3·34? |

* From our own work.

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been repeated often enough to ensure reproducibility, as would be desirable in radiation work. The results and conclusions should therefore be taken as tentative work only, and may I say that I reserve judgment.

Our conclusions are that, with a constant dose rate, peroxide production is proportional to the dose; and the energy yield for peroxide production $G$ with a dose rate of approximately 400,000 rads/hr is about 18. Since peroxide production is a chain reaction this energy yield would be dependent on the dose rate and, as our experiments indicate, the higher the dose rate, the lower the peroxide production per dose: e.g. with the above-mentioned dose rate, a dose of 500,000 rads leads to a peroxide value of 8·6; with a dose rate of about 150,000 rads/hr the same dose leads to a peroxide value of 15·2. In the initial stage of the chain reaction where peroxide decomposition is small the amount of peroxide formed at a constant dose was found to be inversely proportional to the square root of the dose rate. The range of dose rates investigated was 500 to 400,000 rads/hr. This dependence on dose rate is not unusual for chain reactions and would be expected from a purely kinetic point of view in any sequence of reactions where a secondary free radical reaction competes with a secondary radical–radical combination. It appears, therefore, that in food irradiation one should try to apply as high a dose rate as possible to minimize the effect of chain reactions. Indeed the above-mentioned findings with methyl linoleate justify a careful examination and reinvestigation of all the effects at extremely high dose rates, e.g. 1 Mrad/min, and conceivably food irradiation would become a better practical proposition under such conditions. A marked dependence on dose rate was also found in the irradiation of thiourea by DALE and DAVIES[14] (Fig. 1 is taken from their work). Their very high $G$ values are explicable only on the basis of a chain reaction. Returning to Table 1 we note that carbonyl formation is not as strictly proportional to dose as peroxide formation. This may be due partially to peroxide decomposition but also partially

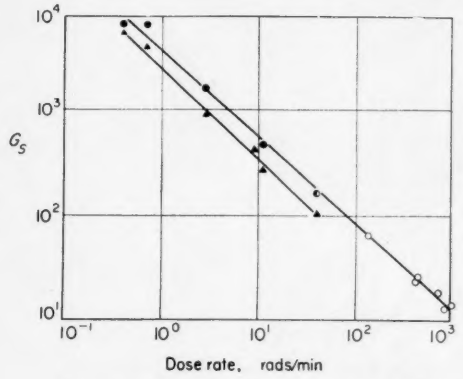


FIG. 1. Radiation effects on thiourea solutions. Relationship between $G_s$ and dose rate. ○ total dose $2{\cdot}7 \times 10^5$ rads; ● total dose $4 \times 10^4$ rads; ▲ total dose $10^4$ rads; ◐ result of two experiments, one with total dose $4 \times 10^4$ rads and the other with total dose $2{\cdot}7 \times 10^5$ rads.

Reproduced from a paper by DALE W. M. and DAVIES J. V. in *Rad. Res.* **7**, 35 (1957).

to a certain lack of accuracy in carbonyl determinations. During recent years the spectrophotometric method developed by HENICK *et al.*[15] has been widely used and we have experimented with it but finally arrived at the conclusion that for methyl linoleate and linoleic acid the method of LAPPIN and CLARK is superior.[16] The main difference is that, in the latter, the reaction with dinitrophenyl hydrazine is carried out in methyl alcohol instead of benzene and peroxide decomposition during the determination is much less significant in methyl alcohol due to its higher polarity. However, blank tests were sometimes so high that it was considered wiser to mark certain results with question marks. We do note that practically all free radical acceptors reduced peroxide formation but so far the results obtained with our new flavones are not so encouraging as in the normal course of oxidative rancidity. Nevertheless, it should be noted that it is not certain that the specified antioxidant concentration was in fact reached with the flavones and continuous redissolution as would be the case in shaking Warburg vessels has not yet been attempted. When I said that practically all antioxidants reduced peroxide

TABLE 2

| No. Substance | Protection for polymer (%) | Number of mice surviving out of 10 |
|---|---|---|
| Amino Compounds–Cont. | | |
| 28. *m*-Tyramine | 71 | 10 |
| 29. Tryptamine | 76 | 10($1 \times 10^{-5}$ mole) |
| Hydroxy Compounds | | |
| 30. Methanol | 27 | 1 |
| Ethanol | 39·2 | |
| *n*-Propanol | 51 | |
| *Iso*propanol | | 0 |
| 31. *n*-Butanol | 49 | 0 |
| *n*-Amyl alcohol | 44 | |
| Ethylene glycol | 22 | |
| Propylene glycol | | 8($8 \times 10^{-4}$ mole) |
| 32. Glycerol | 12 | 0 |
| Trimethylene glycol | 36 | |
| 33. Lactic acid (Na salt) | 14 | 0 |
| Glucose | 21 | |
| 34. Glucose | 47($16 \times 10^{-4}$ M) | 0–3($1{\cdot}5 \times 10^{-3}$ mole) |
| 35. Fructose | 40 | |
| Fructose | 61($16 \times 10^{-4}$ M) | 8($1{\cdot}5 \times 10^{-3}$ mole) |
| 36. Salicylic acid (Na salt) | 48 | 5 |
| 37. Phenol | 63 | 4($2 \times 10^{-5}$ mole) |
| Hydroquinone | 78 | Toxic for all mice |
| Resorcinol | 25 | 3 (out of 4) |
| Hydrocarbons and Unsaturated compounds | | |
| *n*-Hexane | 3 | |
| *Cyclo*hexane | 33 | |
| *Cyclo*hexene | 23 | |
| Methylpentadiene | 22 | |
| Benzene | 45 | |
| Dibenzanthracene | 51($4 \times 10^{-4}$ M) | |
| *Cyclo*octatetraene | 14 | |
| Methacrylonitrile | 76 | |
| Methacrylamide | 45 | |
| Methacrylic acid (Na salt) | 39 | |
| Maleic acid (Na salt) | 39 | |
| Oleic acid (Na salt) | 15 | |
| Allyl alcohol | 68 | |
| 38. Allylamine | 60 | 0 ($10^{-5}$ mole) |
| Divinyl sulfone | 53 | |
| Nitriles | | |
| 39. Sodium cyanide | 80($4 \times 10^{-4}$ M) | 5–7 ($2 \times 10^{-6}$ mole) |
| Acetonitrile | 0 | |
| 40. Malononitrile | 38 | 8 ($2 \times 10^{-6}$ mole) |
| Methacrylonitrile | 76 | |
| Allyl cyanide | 78 | |

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formation, one notable exception is carboxymethylmercaptosuccinic acid, which had no effect. On the other hand, diphenyl *p*-phenylenediamine proved to be the best of the free radical acceptors tested so far. This may be a general phenomenon for many amino compounds and perhaps future studies should be devoted to a greater extent to the exploration of certain amino derivatives, particularly those with a basic quinoid structure, e.g. the one mentioned, diphenyl *p*-phenylenediamine. The work to be done appears to be of very large scope: we have to explore all the kinetic detail of concentration and dose rate dependence, fate of free radical acceptor molecules and perhaps only when we gain more insight into the question as to why a particular free radical acceptor is better than another, shall we be able to make a great leap forward, the desirable and necessary quantum jump. The difference in radiation protection obtained with pentahydroxyflavones is not sufficiently large to be of great encouragement and only when we succeed in making the more fat-soluble long chain alkyl derivatives would there be, so it appears, real hope of further progress. Such work is in hand but we also must pay increased attention to amino compounds, and although the result with carboxymethylmercaptosuccinic acid is disappointing, other SH derivatives will also be examined.

A large number of protective reagents were examined in a different context by ALEXANDER *et al.*[17] In this investigation, a large number of protective substances were compared in their effect on the degradation of polymers, on the one hand, and the survival of mice subjected to radiation, on the other. It is very difficult to find rhyme and reason with regard to a systematic analysis of the protective efficiency of all the compounds. Part of this selection is given in Table 2. Again it is noteworthy that some of the amino compounds were particularly efficient protective reagents but it is quite likely that many of these substances react through energy transfer.

Finally, I should like to discuss briefly the work of LUNDBERG *et al.*,[7] the results of which were published in 1957. Our approach is, to some extent, related to theirs but, while LUNDBERG and his co-workers were content to examine the effect of a single antioxidant, we specialize in the line of free radical acceptors and it is in this field that we hope to make our contribution. Table 3 presents a survey of results obtained by LUNDBERG and his co-workers with various fatty acids and their esters. As expected, with methyl linoleate a much higher peroxide value is obtained than with methyl oleate or methyl palmitate. Even here it is seen that reproducibility is not too good. Furthermore, it can be learned that in the absence of oxygen, peroxide decreases but carbonyl values and flavour scores appear to be unaffected (the score varying from zero to three with decreasing acceptability). In general, I consider that the carbonyl values reported here are too high and this may be due to the method used, which, I think, may well err on the high side. The danger of judging by flavour score is perhaps well illustrated in Table 4 where an irradiated sample with a peroxide value of 68 and a very high carbonyl value was reported to have a rather acceptable flavour score of 0·9, better than the flavour score of a sample with a peroxide value of 18·5. Needless to say, the authors were not themselves prepared to accept the validity of this flavour score.

The effect of propyl gallate is shown in Table 5. This was the only antioxidant used by LUNDBERG and his co-workers. The effect on the peroxide value is rather better than we experienced; on the other hand, the effect on carbonyl formation is rather worse. Some results are also reported which show the effect of water and emulsifying reagents in general, leading to a reduction in peroxide formation.

Table 6, also taken from the work of LUNDBERG and his co-workers, indicates that irradiation of methyl linoleate at a low temperature is no solution. The peroxide values are, in fact, higher. This is in agreement with the experience of Hannan in the irradiation of butter.

We have ourselves carried out a number

TABLE 3. Irradiation with and without oxygen
($2 \times 10^6$ rep, ambient temperature)

| Substrate | Peroxide value (mmole/kg) | Carbonyl compounds (mmole/kg) | | Flavor score |
|---|---|---|---|---|
| | | Unsaturated | Saturated | |
| | Under oxygen ($\beta$ or $\gamma$-rays) | | | |
| Methyl palmitate | 2·2 | 0·3 | 4·9 | 0·7 |
| | 2·3 | 0·6 | 5·0 | 1·8 |
| | 3·2[a] | 0·5 | 4·3 | 1·8 |
| | 2·8[a] | 0·7 | 3·6 | 2·0 |
| Methyl oleate | 4·6 | 1·8 | 3·7 | 1·4 |
| Methyl linoleate | 12·8 | 1·8 | 2·8 | 0·9 |
| | 6·9 | 3·1 | 3·6 | 1·3 |
| | 18·5 | 5·4 | 5·1 | 1·7 |
| Corn oil methyl esters | 19·4 | 9·3 | 3·5 | 2·2 |
| | 11·4 | 8·7 | 3·8 | 1·2 |
| | 19·6 | 9·3 | 5·8 | 1·3 |
| Lard | 10·1[a] | 3·0 | 9·8 | 2·1 |
| | 10·6[a] | 2·8 | 10·1 | 1·7 |
| | Under nitrogen ($\beta$ or $\gamma$-rays) | | | |
| Methyl palmitate | 1·1[a] | 0·1 | 2·0 | 0·5 |
| | 1·8[a] | 0·2 | 2·1 | 1·0 |
| Methyl linoleate | 0·7 | 1·2 | 2·6 | 1·7 |
| | 0·7 | 1·9 | 1·7 | 1·0 |
| Corn oil methyl esters | 1·8 | 6·2 | 1·5 | 1·7 |
| Lard | 1·7 | 1·6 | 6·8 | 0·7 |
| | Under vacuum ($\gamma$-rays) | | | |
| Methyl palmitate | 0 | 0·2 | 1·9 | 0·5 |
| | 0[a] | 0·1 | 2·0 | 0·5 |
| | 1·0[a] | 0·2 | 2·0 | 0·5 |
| Methyl linoleate | 10·8[b] | 1·2 | 2·3 | 1·6 |
| Lard | 0·5[a] | 1·4 | 5·9 | 0·9 |
| | 0·5[a] | 1·4 | 6·3 | 1·3 |

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[a] Obtained from duplicate samples irradiated at the same time. Other values obtained in other experiments.
[b] Value unusually high and considered in error.

TABLE 4. Effect of multiple $\beta$-ray doses on methyl linoleate under oxygen

| Irradiation conditions | | Peroxide value (mmole/kg) | Carbonyl compounds (mmole/kg) | | Flavor score |
|---|---|---|---|---|---|
| Dose (rep) | Temp.(°C) | | Unsaturated | Saturated | |
| One $2 \times 10^6$ | 28 | 18·5 | 5·4 | 5·1 | 1·7 |
| Ten $2 \times 10^5$ | 28 | 68·0 | 14·4 | 13·2 | 0·9 |
| One $2 \times 10^6$ | −78 | 23·0 | 3·7 | 6·8 | 1·0 |
| Ten $2 \times 10^5$ | −78 | 32·3 | 6·2 | 5·2 | 2·2 |

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TABLE 5. Effect of propyl gallate on irradiation with oxygen
($\gamma$-rays, $2 \times 10^6$ rep, ambient temperature)

| Sample | Peroxide value, mmole/kg | Carbonyl compounds, mmole/kg | | Flavor score |
|---|---|---|---|---|
| | | Unsaturated | Saturated | |
| Methyl palmitate | 2·2 | 0·3 | 4·9 | 0·7 |
| + 0·01% P.G. | 0·1 | 0·2 | 1·9 | 0·3 |
| + water + dextrin | 0·3 | 0·3 | 2·1 | 1·5 |
| + water + dextrin + 0·01% P.G. | 0·3 | 0·3 | 2·1 | 1·2 |
| Methyl linoleate | 12·8 | 1·8 | 2·8 | 0·9 |
| + 0·01% P.G. | 2·6 | 0·8 | 3·0 | 0·9 |
| + water + dextrin | 6·9 | 0·9 | 2·4 | 1·4 |
| + water + dextrin + 0·01% P.G. | 3·0 | 0·7 | 2·3 | 0·9 |
| Lard | 10·1 | 3·0 | 9·8 | 2·1 |
| | 10·6 | 2·8 | 10·1 | 1·7 |
| + 0·01% P.G. | 7·1 | 2·7 | 8·2 | 2·0 |
| + 0·01% P.G. | 6·8 | 2·6 | 8·3 | 1·4 |

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TABLE 6. Effect of temperature of irradiation
($\beta$-rays, $2 \times 10^6$ rep, oxygen atmosphere)

| Sample | Irradiation temp. (°C) | Peroxide value (mmole/kg) | Carbonyl compounds (mmole/kg) | | Flavor score |
|---|---|---|---|---|---|
| | | | Unsaturated | Saturated | |
| After 3·5 hr at 25°C | | | | | |
| Methyl oleate | −20 | 14·60 | | | |
| | −35 | 12·90 | | | |
| Methyl linoleate | −45 | 42·90 | | | |
| | −55 | 53·40 | | | |
| After 3·5 hr at −78°C | | | | | |
| Methyl oleate | −20 | 16·30 | | | |
| | −35 | 16·10 | | | |
| Methyl linoleate | −45 | 19·00 | | | |
| | −55 | 26·30 | | | |
| After 3 days at −78°C | | | | | |
| Methyl oleate | 28 | 4·6 | 1·8 | 3·7 | 1·4 |
| | −20 | 4·3 | 1·6 | 4·6 | 0·7 |
| | −78 | 5·3 | 2·8 | 5·0 | 0·6 |
| Methyl linoleate | 28 | 18·5 | 5·4 | 5·1 | 1·7 |
| | −20 | 15·8 | 5·2 | 4·9 | 1·6 |
| | −20 | 18·0 | 4·7 | 4·9 | 1·7 |
| | −78 | 14·1 | 3·8 | 4·6 | ... |

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of experiments in emulsions where we had some success with partially alkylated hydroxyflavones. Some of these results were reported at last year's meeting in Cambridge. Since then we have carried out further experiments with emulsions and also with milk, but this would really take me a little too far away from the fundamental side to the applied side. Mr. Horne will be speaking about milk in detail. I should only like to point out that amongst the conventional antioxidants which were recently allowed in this country we found that BHA emulsified in milk gave the best protection.

As far as new free radical acceptors are concerned, studies will have to be undertaken to establish their lack of toxicity, even though they may be closely related to naturally occurring compounds which are consumed in fair quantities. Another problem amongst the many perplexing aspects is the question of whether efficient free radical acceptors would not protect micro-organisms with equal efficiency. I do not think that this would, in fact, happen, but it will have to be demonstrated in every individual case. Conceivably some free radical acceptors would be converted during irradiation into quinones with antibiotic properties and thus automatically increase the lethal effect on micro-organisms. One might call them "radio-antibiotics", a term not yet found in the literature.

It is, I believe, too early to prophesy whether food irradiation can be successfully applied, but perhaps a great deal might depend on the possibility of discovering an ideal free radical acceptor. Undoubtedly, we have not yet reached the end of the beginning of our research.

*Acknowledgement*—I should like to express my thanks to Miss M. R. Menzies for her valuable assistance in this work.

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# Free Radical Investigations in the United States

G. TRIPP*

Quartermaster Food and Container Institute for the Armed Forces, Chicago, Ill., U.S.A.

THE work in the United States has mainly been directed towards effects of radiation on whole foods. Extensive basic studies such as those reported by Professor WEISS and Dr. URI have received little attention.

Limited studies in the U.S. have been in progress using paramagnetic resonance to determine the amount of free radical formation, types of radicals formed and to determine the ultimate fate of the free radical. These studies have been conducted with dry foods, moist foods at ambient conditions and frozen moist foods. In the case of whole foods at ambient temperatures, the free radicals are formed and dissipated by reactions in the food. When dry foods or frozen foods are irradiated, free radicals are formed and persist for long periods. Upon thawing the frozen food or moistening the dry food, the free radicals enter into reaction with the food and are dissipated. For example, irradiation of crystalline sucrose produced free radicals in the sugar. After several months of storage the free radicals were still present. Solution of the sucrose in water resulted in reactions between the free radicals and other constituents of the solution. The persistence of free radicals in the frozen food explains, in part, why greater differences have not been observed between foods irradiated in the frozen state as compared to the same food irradiated at ambient temperatures. A similar situation exists with polymeric materials. We have observed that free radicals are formed in solid polyethylene which persist for long periods. Melting of the irradiated material permits further reaction of the free radical.

Studies have been conducted to determine if the effects of free radicals could be minimized through use of free radical acceptors. Numerous substances have been evaluated including ascorbic acid, tocopherols, propyl gallate, cysteine and amino compounds. Results of these studies, although not highly successful, indicate this approach may be of value and requires more study. Addition of tomato products to meats and acidification of meats with acetic acid has proved highly beneficial in preventing off-odor and flavor development in beef.

Dr. URI has referred to the *G* values of various free radical reactions. Recent reports in the literature indicate that in the irradiation of sodium nitrate and nitrite in the solid state or as a solution the *G* value is related to the ratio of the absolute volume of the molecule to the apparent volume being irradiated. Further, in work with teflon, we have observed that the quantity of fluorine released is related to the absolute volume of fluorine present to the apparent volume of the solid. Additional studies along these lines may aid in understanding the chemical changes occurring during irradiation of food and other substances.

Dr. URI has referred to the possible influence of dose rate. We are in sympathy with these views and are currently reconsidering the possible influence of dose rate on micro-organisms and food. In our earlier work, we had concluded that a dose rate effect did not exist; however, the inadequacies of methods of measuring dosage during this early work requires that we re-examine this possibility.

*FAO Consultant.

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# Discussion

Dr. ROBERTS (U.K.): Referring to the results of Dr. Uri on the effect of temperature on the reactions, in Table 4 of his paper, can these reactions be explained completely on a free radical basis? Ion–molecule reactions may also play a part.

Dr. URI (U.K.): A free radical explanation seems the most likely for these particular reactions.

Professor MANIL (Belgium): I should like to ask Professor Weiss if the organic peroxides to which he referred are decomposed by catalase.

Professor WEISS (U.K.): Only very few of the new hydroperoxides produced by irradiation have been investigated from this point of view, up to the present. However, we do know that thymine-hydroxy-hydroperoxide is not decomposed by catalase.

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# Chemical Changes Produced in Lipids by Irradiation

B. COLEBY

Low Temperature Station for Research in Biochemistry and Biophysics,
Department of Scientific and Industrial Research, Cambridge

Irradiation gives rise to off-flavours in lipids which, with some foods, are a serious disadvantage. The off-flavours show a resemblance to those encountered in oxidative rancidity, and experiments with model systems have shown that the course of reactions is somewhat similar in the two cases, i.e. hydroperoxides and carbonyl compounds are formed. Usually, the molecule is attacked in a limited number of places; and this has also been observed with some phospholipids. Removal of oxygen during irradiation and storage prevents some, but not all, of the changes. Antioxidants present in natural lipids can profoundly influence the reactions, and the physical state of the lipid, determined by temperature, can play an important role during both irradiation and subsequent storage.

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GENERALLY speaking, the lipid constituents of many foods are less susceptible to microbial spoilage than other major components. Nevertheless, prolonged storage can often lead to the deterioration of lipids through oxidative chemical attack, and this may be the factor limiting storage life for foods protected from microbial spoilage, e.g. frozen meat.[1]

The initial step of oxidation is the formation of hydroperoxides, which degrade to the carbonyl compounds responsible for the odours and flavours of rancid fat. A similar type of chemical behaviour is often observed during the irradiation and subsequent storage of many lipids. In some foods, such as meats, the dominant irradiation odours and flavours do not appear to arise principally in the lipid phase; but in other foods, particularly dairy commodities, some of the flavours induced by irradiation resemble quite closely those encountered in oxidative rancidity. It is known that if meat fat is minced with lean tissue, the irradiation induced flavours are less severe;[2] and there is evidence that carbonyl compounds can react with the characteristic sulphur compounds formed by the irradiation of lean meat.[3] There are good reasons, therefore, for wishing to understand the chemical effects of radiation on lipids.

Chemical investigation of lipids is notoriously difficult. Several fatty acids can usually be isolated from any given fat, and even the individual triglyceride molecule commonly contains more than one fatty acid residue. Variations in the chain length and degree of unsaturation of the fatty acids produce a group of compounds of closely similar physical properties, which is difficult to fractionate. Radiation chemical studies usually have the added difficulty of being concerned with small amounts of chemical change. In the circumstances it is not surprising that we lack detailed knowledge of the radiation chemistry of lipids; and what we do know is often gleaned from experiments with model systems.

In the radiation chemistry of aqueous systems, it is well known that much of the energy is absorbed by the water, forming free radicals; and subsequent changes in a solute can often be explained in terms of the reactions of hydrogen atoms, hydroxyl radicals, hydrogen and hydrogen peroxides.[4] It is understandable that quite specific chemical reactions may be observed. In the cases of lipids, there is no solvent, so

that reactions cannot be explained in terms of the formation of a limited number of radical species. Since the most of the mass of the triglyceride molecule is in the fatty acid side chains, a corresponding proportion of the radiation energy must be absorbed in these parts of the molecule. In spite of this, however, there is enough evidence to show that chemical changes induced by irradiation can be specific in nature, being usually located near reactive groupings in the lipid molecules. Obviously some type of energy transfer must occur.

If a saturated hydrocarbon is irradiated, there is rupture of C–H and C–C bonds, giving free radicals. In the absence of oxygen, these can react in a number of ways: dehydrogenation, chain rupture and dimerization are commonly observed.[5] Apparently there is little preference for any particular bond to be broken. If, on the other hand, a substance containing a reactive group in the hydrocarbon chain is irradiated, then, in many cases, reaction occurs quite specifically at the reactive centre. One or two illustrations will suffice: alkyl iodides rupture almost exclusively at the C–I bond on irradiation;[6] and there are many examples in the irradiation of plastics, such as the preferential disappearance of the vinylidene groupings in polyethylene.[7] Reactive groups tend to react in characteristic ways, an important case being the tendency of ethylenic linkages to lead to polymerization.[5]

Fatty acids also undergo one principal reaction, that of decarboxylation, to give carbon dioxide and the parent hydrocarbon.[8,9] Decarboxylation seems to be largely unaffected by the chain length of the acid, but minor reactions occur too, yielding carbon monoxide, small hydrocarbons, some unsaturated products, and water. With unsaturated fatty acids, polymerization is the major reaction, but some decarboxylation can still occur.[10]

In the presence of oxygen, rather different reactions occur. The principal effect of irradiation appears to be to induce reactions very similar to those occurring during autoxidation, i.e. hydroperoxide formation leading to carbonyl production.[11] Autoxidation studies indicate that hydroperoxide formation occurs in only a limited number of places: e.g. with methyl linoleate, attack is on carbon atoms 9 and 13;[12] and with methyl oleate attack takes place on carbon atoms 8, 9, 10 and 11, mainly in the last two positions. In the case of methyl oleate such attack has been observed during both autoxidation and irradiation, and similar carbonyl compounds were isolated in the two cases.[13]

Another example of a lipid undergoing specific reactions on irradiation is in the case of lecithin and hydrogenated lecithin.

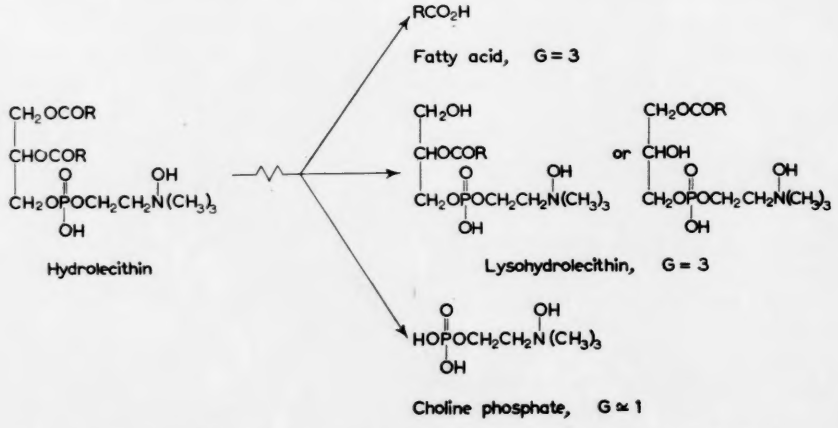


FIG. 1. The degradation of hydrogenated lecithin by high energy electrons (2 MeV).

When hydrogenated lecithin is irradiated *in vacuo*, the ester linkages are split, giving a lyso derivative, fatty acids and choline phosphate as products, as shown in Fig. 1. The yields show that virtually every ionization is effective in breaking an ester bond, and the molecular weight (300) of the liberated fatty acids shows that they do not undergo degradation during cleavage. In the presence of oxygen, hydrogenated lecithin gives the same products but in reduced yield (e.g. fatty acids are liberated with $G \sim 1{\cdot}5$), and in addition hydroperoxides are formed. Again, the yield of hydroperoxides approximates to one molecule of peroxide from each ionization. Natural lecithin shows some differences in behaviour, owing to the ethylenic linkages in the molecule. As a rule, egg lecithin contains an average of about two such linkages per molecule, and practically all of the unsaturated acids are located on the α-carbon of the glycerol.[14] Several acids of varying degrees of unsaturation may be isolated from a given sample of lecithin; and usually the largest acids ($C_{20}$ and $C_{22}$) are the most unsaturated, having 3, 4 or more ethylenic bonds per molecule.[15] After irradiation, the isolated fatty acids are found to be of rather higher molecular weight than the average (360 as against 280), and to be mono-unsaturated, so that partial hydrogenation must occur during the preferential cleavage of the longer fatty acids. Some hydrogenation of the lecithin takes place during irradiation, the *G*-value for this reaction being ~2.

Not surprisingly, information about the chemical effects of irradiation on natural lipids is much less precise. Natural lipid systems usually contain minor constituents such as pigments or natural antioxidants, and these profoundly modify the radiation chemical behaviour. For the most part, observations have been confined to the measurement of gross chemical changes such as the formation of hydroperoxides, carbonyl compounds, free fatty acids, polymers and changes in unsaturation. The presence or absence of oxygen plays a decisive role in these changes. Usually, each fat behaves in a characteristic fashion.[16] Much depends upon the origin of the lipid: vegetable oils are often more resistant than animal fats, presumably due to the higher content of natural antioxidants.

The observed reactions can usually be interpreted on the assumption that free radicals are formed; and if the lipid is of a suitable nature, these radicals can persist for long periods, causing changes after the act of irradiation. The work of HANNAN[17] illustrates this. If butterfat is mixed with

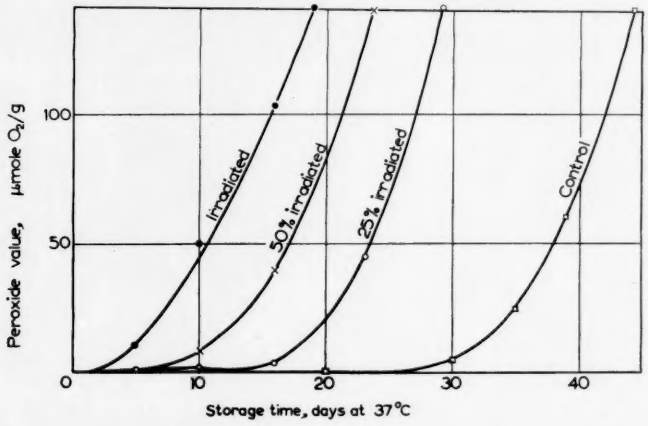


FIG. 2. Autoxidation of a typical sample of irradiated butterfat compared with that of the control fat and various mixtures of the two.[17]

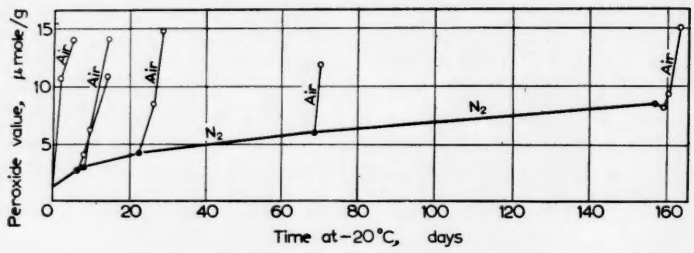


FIG. 3. Peroxide values of butterfat irradiated with 2 MeV electrons ($10^6$ rads) and stored at −20°C in nitrogen for various periods and then in air.[17]

irradiated butterfat in various proportions, the lag period before the onset of autoxidation is reduced in proportion to the amount of irradiated material added (Fig. 2). Presumably this is due to the reaction of radicals with natural antioxidants in the unirradiated butterfat. Similarly, if the fat is irradiated in the absence of oxygen, little hydroperoxide is formed, but admission of oxygen after various periods of storage immediately causes hydroperoxide formation (Fig. 3). Most workers agree that the removal of oxygen from lipids prevents both the development of hydroperoxides and the production of off-flavours on irradiation. But the reactions are undoubtedly complex, since carbonyl formation has been reported even in the absence of oxygen.[18]

The physical state of the lipid has an important influence upon the reactions, probably by determining the life of the radicals formed by irradiation. Experiments with butterfat provide a good illustration.[17] A small proportion of hydroperoxide is formed during irradiation, but the condition which determines whether hydroperoxides develop after irradiation is whether the radicals are allowed to persist. Their life can be terminated by warming the fat to a liquid or semi-liquid state when they react without hydroperoxide formation. If the fat is stored in a solid state at lower temperature, the radicals persist and react with oxygen as it diffuses into the fat, giving hydroperoxides. The optimum temperature for the formation of hydroperoxides is about −30°C. This is shown in Fig. 4.

It will be evident that much study is

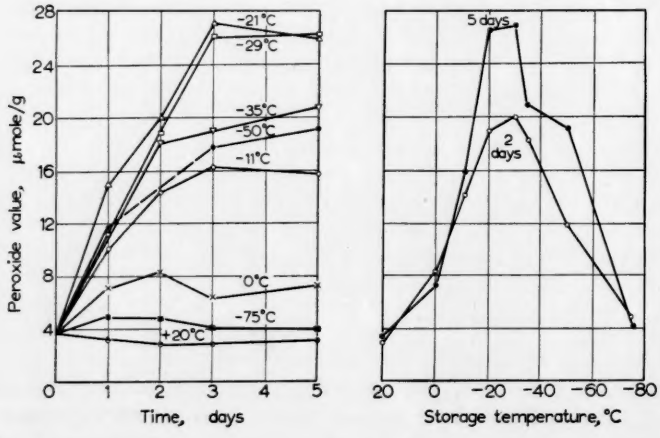


FIG. 4. The effect of storage at various temperatures on the peroxide value of butterfat irradiated with 50 kV X-rays ($10^6$ rads) at −70°C.[17]

necessary to understand more fully the radiation chemistry of lipids; and since each behaves in its own characteristic fashion, individual fats will need to be investigated.

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# The Effects of Radiation on Proteins

B. S. SCHWEIGERT

American Meat Institute Foundation and
Department of Biochemistry, The University of Chicago, Chicago, Illinois, U.S.A.

MARKED physical and biological changes have been noted in proteins subjected to treatment with ionizing radiations in spite of the fact that relatively few of the chemical bonds in the materials irradiated have been broken. The major chemical changes noted include denaturation, degradation and polymerization. The allergenicity of proteins has been shown to be particularly sensitive to irradiation. One of the primary changes noted after the irradiation of proteins is their marked sensitivity to temperature changes of the stored solutions. We have noted this with irradiated solutions of the meat pigment myoglobin, a conjugated protein, in which warming of solutions to room temperature results in extensive precipitation of the protein, presumably due to aggregation.

Other studies show increases in viscosity of protein solutions and a decrease in solubility. Irradiated solutions of casein, for example, will form an insoluble gel. The primary chemical change appears to involve the sulfhydryl bond in the protein and possibly the phenolic group of tyrosine. The oxidative effects on the sulfhydryl groups result in polymerization through disulphide bonds.

Studies on the allergenicity of milk proteins indicate a marked reduction in this property during irradiation with dosages up to 9·3 Mrads, with appreciable changes noted about 2·8 Mrads. The results also suggest that considerable amounts of immune globulin and other protein fractions undergo denaturation and fragmentation to form polymerized material which is non-mobile during electrophoresis. Ovalbumin shows a greater loss in serologic activity due to irradiation than can be accounted for by the amount of protein denatured as determined by precipitation of the protein under standard conditions.

The presence of salts and reduced temperature during irradiation protect proteins, such as serum and egg albumin, from irradiation damage. Treatment with 100,000–200,000 rads will show some evidence of dimer formation with these proteins. This can be prevented by the addition of cystine.

The activity of enzymes due to irradiation has received a great deal of attention because of the importance of these substances to the quality of fresh foods that have been stored for varying periods of time. Enzymes are relatively resistant to inactivation by irradiation. In most cases inactivation requires over 5 Mrads, while many proteolytic enzymes require 12 to 70 Mrads treatment. The proteolytic enzyme activity of beef muscle is reduced approximately 50 per cent with a dosage of 1·6 Mrads. Sulfhydryl enzymes are more sensitive to irradiation. As suggested by the above comments, enzymes are less stable in pure solutions than in complex mixtures or in the dry state. The enzyme, catalase, is inactivated by 25,000 rads in solution, and in potatoes by 5 Mrads.

Irradiation of protein constituents, such as individual amino acids or peptides, also reveals a number of important changes. The main changes that occur include deamination, decarboxylation, and the formation of hydrogen sulfide and methyl mercaptan from the sulfur containing amino acids. If the meat pigment, myoglobin, is irradiated in the presence of cysteine, considerable



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quantities of the green pigment, sulfmyoglobin, are formed. Undesirable odors, for example, have been obtained by irradiation of leucine and the major product formed was *iso*valeraldehyde. Irradiation of glycine in the presence of a sugar, such as glucose, results in the formation of brown polymers. The irradiation of glycylglycine results in ammonia liberation and breakage of the peptide linkage. Further studies have shown that there is some destruction of several amino acids with high dosages (17 Mrads). Little, if any, destruction has been noted with sterilization dosages, however, of amino acids when present in intact proteins. Thus, we may conclude there is no major loss in amino acid content but profound changes in the physical and biological characteristics of proteins that have been irradiated.

Changes in foods that have been irradiated appear to be partly related to changes in the protein. Meat, for example, appears to have a reduced water-holding capacity, which presumably is due to alteration in the affinity of denatured proteins for water. The degree of inactivation of enzymes perhaps has the greatest influence on the characteristics of irradiated foods. The relative resistance of these substances to irradiation has prompted increased attention to precooking foods, such as meat, prior to irradiation treatment to avoid the autolytic changes that occur during storage with irradiated foods.

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# The Chemical Action of Gamma Radiation on Aqueous Solutions of Carbohydrates

G. O. PHILLIPS and G. J. MOODY
University College, Cardiff

Using paper chromatographic and radioactive tracer methods, the products formed when aqueous solutions of carbohydrates are irradiated with ionizing radiations are identified and quantitatively estimated.

In dilute aqueous solutions D-glucose is degraded to D-glucuronic acid, D-gluconic acid, glyoxal, D-arabinose, D-erythrose, formaldehyde, saccharic acid and 1:3-dihydroxyacetone, which have been estimated quantitatively. By following the yield–dose curves for the main products, the primary and secondary products are distinguished, and the main types of degradation identified.

The main products formed during the irradiation of aqueous dextran solutions are glucose, *iso*maltose, *iso*maltotriose, gluconic acid, glucuronic acid, glyoxal, erythrose and glyceraldehyde. These products are considered to arise from at least two independent processes.

The main attack when D-sorbitol solutions are irradiated with $\gamma$-rays is located at the primary alcohol groups in the molecule. The primary products are D-glucose and L-gulose, and appreciable amounts of D-arabinose and L-xylose were formed by secondary attack. A possible mechanism for the process is suggested.

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EXTENSIVE chemical changes have been induced in organic molecules by exposure to ionizing radiations.[1–10] The changes which occur during the irradiation of carbohydrates, however, have received little detailed study, and attention appears to have been focused only on the physical changes observable during irradiation, such as pH, optical rotation, reducing power, viscosity and ultraviolet absorption spectra.[11–15] In this paper we consider the chemical action of ionizing radiations on aqueous solutions of D-glucose, dextran and D-sorbitol. Paper chromatographic and radioactive tracer methods were used for the identification and estimation of the main products in order to ascertain the main steps of the degradation process. Full experimental details have been described.[16–18] The cobalt-60 source (200 c) employed for the gamma irradiations was of similar design to that developed at A.E.R.E., Harwell, and described by GIBSON and PEARCE.[19]

## 1. ACTION OF GAMMA RADIATIONS ON AQUEOUS SOLUTIONS OF D-GLUCOSE

D-Glucose in aqueous solution undergoes extensive degradation under the action of ionizing radiations.[6] Chromatography of the solutions irradiated with gamma radiation indicated the presence of a variety of constituents; the main organic products are listed in Table 1.

### *Estimation of products by isotope dilution method*

The isotope dilution method applied directly to the irradiated mixture confirmed the results obtained by paper chromatography. By this method further products were also identified. A typical estimation is described in full. An aqueous solution (100 ml) of glucose (11·07 mmoles) of specific activity 2·8 $\mu$c/mmole was irradiated at a dose rate of $2{\cdot}13 \times 10^{17}$ eV min$^{-1}$ ml$^{-1}$ in oxygen for 46·5 hr. The isotope dilution method was applied directly to the untreated irradiated solution.

*Glucose.* The irradiated solution (10 ml)

TABLE 1. Constituents in D-glucose solutions after irradiation

| Method | Irrigant | $R_F$ | |
|---|---|---|---|
| I Paper chromatography | Butan-1-ol–acetic acid–water | 0·11 | Glucuronic acid |
| II Paper chromatography | Butan-1-ol–acetic acid–water | 0·18 | Glucose |
| III Paper chromatography | Butan-1-ol–acetic acid–water | 0·22 | Arabinose |
| IV Paper chromatography | Butan-1-ol–acetic acid–water | 0·34 | Glucurone |
| V Paper chromatography | Butan-1-ol–acetic acid–water | 0·48 | Tetrose |
| VI Autoradiography | Butan-1-ol–acetic acid–water | 0·75 | |
| VII Anilide method[12] | Water–benzene–acetone | 0·52 | Gluconic acid |
| VIII Circular paper chromatography of osazones[13] | Toluene–ethanol–water | 0·97 | Glyoxal |
| V Circular paper chromatography of osazones[13] | Toluene–ethanol–water | 0·52 | Tetrose |

was freeze-dried and any trace of water removed *in vacuo*. Carrier D-glucose (1·37 mmoles), acetic anhydride (1 ml), and fused sodium acetate (200 mg) were added. The mixture was kept at 100° for 2·5 hr, and the penta-O-acetate was then precipitated by water (5 ml). After nine recrystallizations from ethanol, the penta-O-acetylglucose had m.p. 135° and a constant specific activity 0·952 $\mu$c/mmole.

*Glucuronic acid.* Carrier D-glucuronolactone (0·42 mmole) was added to the irradiated solution (5 ml), and the solution reduced to small bulk by distillation. The deposited D-glucurone after five recrystallizations melted at 175° and had constant specific activity 0·35 $\mu$c/mmole.

*Gluconic acid.* The irradiated solution (10 ml) was treated with carrier D-gluconolactone (0·52 mmole) and excess of calcium carbonate. The solution was filtered after 2 days, and the gluconate precipitated with ethanol. Ten recrystallizations gave material of constant specific activity 0·44 $\mu$c/mmole.

*Saccharic acid.* Carrier saccharic acid (0·35 mmole) was added to a portion of the irradiated solution (5 ml). The solution was treated in the usual way, and the deposited acid after six recrystallizations had m.p. 125° and constant specific activity $6{\cdot}8 \times 10^{-3}$ $\mu$c/mmole.

*Formaldehyde.* The distillate obtained during the estimation of glucuronic acid was treated with carrier formaldehyde (0·113 mmole) and 10% dimedone solution (5 ml). After 48 hr the dimedone complex separated and was recrystallized until it had m.p. 189° and constant specific activity 0·02 $\mu$c/mmole.

*Glyoxal.* The irradiated solution (10 ml) was treated with phenylhydrazine (1·5 ml) and glacial acetic acid (1·0 ml). After seven recrystallizations from benzene, glyoxal *bis*-phenylhydrazone was obtained with m.p. 170° and constant specific activity 0·066 $\mu$c/mmole.

*Dihydroxyacetone.* Carrier dihydroxyacetone (1·19 mmole), acetic acid (1 ml), and phenylhydrazine (1·2 ml) were added to the irradiated solution (20 ml) and the mixture was boiled for 5 min. Seven recrystallizations from benzene gave pure D-glycerosazone, m.p. 129°, constant specific activity 0·68 $\mu$c/mmole.

D-*Arabinose.* Attempts to estimate D-arabinose as the tetra-O-acetate were unsuccessful. Accordingly isotope dilution was applied by using the osazone. Carrier D-arabinose (1·04 mmole), acetic acid (1·5 ml), and phenylhydrazine were added to the irradiated solution (10 ml), and the mixture was boiled for 15 min. The product required ten recrystallizations to give pure D-arabinosazone, m.p. 159°, of constant specific activity 0·099 $\mu$c/mmole.

D-*Xylose.* The irradiated solution (10 ml) after freeze-drying was treated with carrier D-xylose (1·88 mmole), sodium acetate (160 mg), and acetic anhydride (1 ml) and

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TABLE 2. Products when aqueous solutions of D-glucose are irradiated with gamma radiation in oxygen

(a) Initial glucose, 5·5 mmole
Energy input, $32{\cdot}6 \times 10^{22}$ eV (volume 115 ml)

| Product | D-Glucose | D-Erythrose | Glyoxal | Formaldehyde | D-Gluconic acid | D-Glucuronic acid |
|---|---|---|---|---|---|---|
| Carrier (mmole) | 1·7 | 0·40 | 1·26 | 0·113 | 1·26 | 0·47 |
| Specific activity ($\mu$c/mmole) | 0·483 | 0·77 | 0·49 | 0·053 | 0·50 | 0·92 |
| Yield (mmole) | 1·13 | 0·22 | 0·96 | 0·147 | 0·63 | 1·01 |

D-Arabinose estimated from paper chromatogram, 0·6 mmole

(b) Initial glucose, 11·07 mmole
Energy input, $5{\cdot}8 \times 10^{22}$ eV (volume 100 ml)

| Product | D-Glucose | D-Erythrose | Glyoxal | Formaldehyde | D-Gluconic acid | D-Glucuronic acid |
|---|---|---|---|---|---|---|
| Carrier (mmole) | 1·37 | 0·68 | 1·13 | 0·113 | 0·52 | 1·04 |
| Specific activity ($\mu$c/mmole) | 0·95 | 0·024 | 0·066 | 0·02 | 0·44 | 0·35 |
| Yield (mmole) | 7·05 | 0·09 | 0·85 | 0·05 | 0·44 | 0·60 |

| | D-Arabinose | Saccharic acid | D-Xylose | Dihydroxy-acetone |
|---|---|---|---|---|
| Carrier (mmole) | 1·04 | 0·35 | 1·88 | 1·19 |
| Specific activity ($\mu$c/mmole) | 0·099 | $6{\cdot}8 \times 10^{-3}$ | $7{\cdot}69 \times 10^{-3}$ | 0·068 |
| Yield (mmole) | 0·45 | 0·016 | 0·06 | 0·31 |

refluxed for 2 hr. Eight recrystallizations from ethanol gave tetra-O-acetyl-$\beta$-D-xylose, m.p. 124°, of constant specific activity $7{\cdot}6 \times 10^{-3}$ $\mu$c/mmole.

*D-Erythrose.* A glucose solution (specific activity 4·00 $\mu$c/mmole (150 ml) was irradiated as above and 10 ml were treated with carrier D-erythrose (0·68 mmole) and phenylhydrazine (2 ml) in glacial acetic acid (1·5 ml). The solid which separated after eight recrystallizations from benzene gave D-erythrosazone, m.p. 157°, specific activity 0·024 $\mu$c/mmole. Table 2 shows the yields of the main products at two energy inputs under fully oxygenated conditions.

*Rate of formation of products*

Two methods were used to measure the rate of formation of the products with energy input during the irradiation of aqueous glucose solutions. First, accurately known amounts of irradiated glucose–$C^{14}$ solution which had received progressively increasing doses of radiation were chromatographed, and the radioactivity of the spots was measured. In the second method the product was estimated directly by isotope dilution at several dose levels. The latter method was adopted only when the product could not be isolated easily as a discrete spot on a paper chromatogram. Fig. 1 shows the relation between the energy input and the rate of formation of glucuronic acid, arabinose, and erythrose formed during the irradiation of glucose–$C^{14}$ solutions. The rate of decomposition of glucose with energy input was also measured. Fig. 2 shows the rate of formation of gluconic acid with energy input as estimated by isotope dilution. This method was also used to obtain the yield/dose

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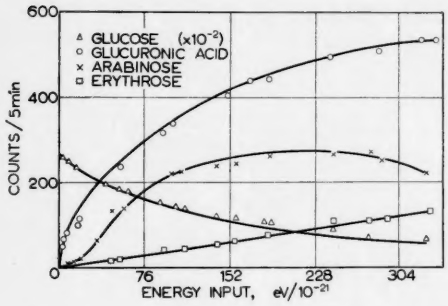



FIG. 1. Rate of formation of products with energy input. Rate of energy input $2{\cdot}13 \times 10^{17}$ eV/min/ml.

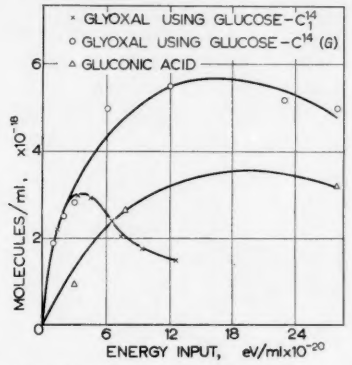



FIG. 2. Rate of formation of glyoxal and gluconic acid with energy input. Rate of energy input $2{\cdot}13 \times 10^{17}$ eV/min/ml.

curve for glyoxal, since it does not move as a discrete spot on paper chromatograms, and gives rise to extensive streaking. Individual experiments were carried out using glucose–$C^{14}$ (*G*) (generally labelled) and glucose–$C_1{}^{14}$, (labelled at $C_1$) in order to ascertain the part of the glucose molecule which is involved in its formation. The results are shown in Fig. 2. The rate of formation of active glyoxal is initially the same from glucose–$C^{14}$ (*G*) and glucose–$C_1{}^{14}$, but at higher dose levels the rate of formation from the latter falls markedly.

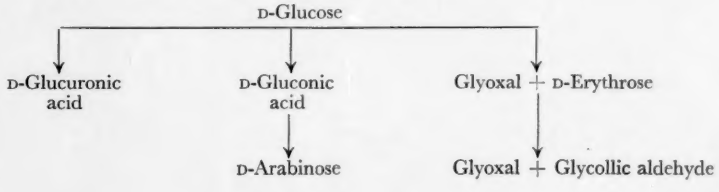
*Pattern of the breakdown*

With oxygenated solutions, the main products formed by the action of gamma radiation are glucuronic acid, gluconic acid, arabinose, glyoxal and dihydroxyacetone. Smaller amounts of formaldehyde, erythrose, xylose and saccharic acid are also present. The estimation of dihydroxyacetone as D-glycerosazone does not serve to differentiate it from D-glyceraldehyde, and the evidence, particularly that provided by the absorption spectra of the irradiated solution, indicates that both are present.[16]

The yield/dose curves for the main constituents enable the primary and secondary products to be distinguished. Glucuronic acid appears to be a primary product because the rate of formation is initially constant, but falls at higher doses until it becomes equal to the rate of degradation.

Similarly, the isotope dilution data for gluconic acid suggest that it is a primary product which again undergoes extensive degradation at high doses. The initial rate of formation of arabinose is low, but increases at the point where the above primary products show appreciable degradation. This behaviour would be expected if arabinose is a secondary product. That glyoxal is a primary product is substantiated by experiments with glucose–$C^{14}$ (*G*) and glucose–$C_1{}^{14}$. The initial rate of formation is the same in both systems, indicating that the $C_1$ and $C_2$ atoms are directly involved in the initial formation of the glyoxal. The amount of glyoxal increases in the experiments using glucose–$C^{14}$ (*G*), whereas with glucose–$C_1{}^{14}$ the concentration reaches a maximum and then falls. It appears accordingly that the glyoxal formed initially from the $C_1$ and $C_2$ atoms in the latter system is effectively diluted by the contribution from the remainder of the molecule.

The main steps in the degradation of D-glucose in aqueous solution, in the presence of oxygen may, therefore, be represented by the following scheme:



There is some evidence from absorption spectra data that the symmetrical scission of the molecule into glyceraldehyde, which partly isomerizes into dihydroxyacetone, may also be a primary process.[16] The secondary product, arabinose is likely to be formed from gluconic acid as a result of the attack of OH radicals.[22]

## 2. DEGRADATION OF AQUEOUS DEXTRAN WITH GAMMA RADIATION

In the present investigation a straight-chain dextran, which can be synthesized by the bacterium lencenostoc, was used.[23] The chemical effects of ionizing radiations on this substance are interesting because it is used as a blood-plasma expander.[24] It serves also as a convenient model polysaccharide linked at $C_6$ in the glucose molecule.

After irradiation the dextran solution was dialysed to effect a separation of the lower saccharides produced. Examination of the dialysate by paper chromatography and electrophoresis indicated that the main products were *iso*maltotriose, *iso*maltose, glucose and erythrose. Streaking on the chromatograms suggested the presence of glyoxal. Treatment of the dialysate with phenylhydrazine gave a solid mixture of osazones, and separation by the circular paper method established the presence of glyoxal *bis*phenylhydrazone, erythrosazone and glucosazone. These osazones were readily separated on a column of alumina[21] by elution with benzene, ether and ethanol–water respectively. The identity of the individual osazones was established by comparing the infra-red and ultraviolet spectra with those of authentic specimens.

The products were also identified and estimated by the isotope dilution method. Dextran–$C^{14}$ diluted with the straight chain dextran under investigation was irradiated in solution. After dialysis to constant rotation the individual products were estimated on the concentrated dialysate.[17] The results for the isotope dilution estimations are shown in Table 3.

By reference to the above pattern of degradation of glucose it is possible to rationalize the results which have been quoted for irradiated dextran solutions. One of the main processes involved is hydrolytic cleavage to form glucose, *iso*maltose and *iso*maltotriose. Estimations of glucose on the basis of isotope dilution with glucosazone and penta-O-acetyl-$\beta$-D-glucose are in fair agreement, and indicate that this monosaccharide is a major product. Two other major products are gluconic and glucuronic acids. The detailed yield/dose curves for the formation of acids and reducing substances indicate that they are both primary processes.[17] It is evident, therefore, that dextran undergoes degradation by at least two independent processes involving scission at the 1–6 link of the molecule. One leads to glucose and lower saccharides, and the other to gluconic and glucuronic acids as represented in the scheme:

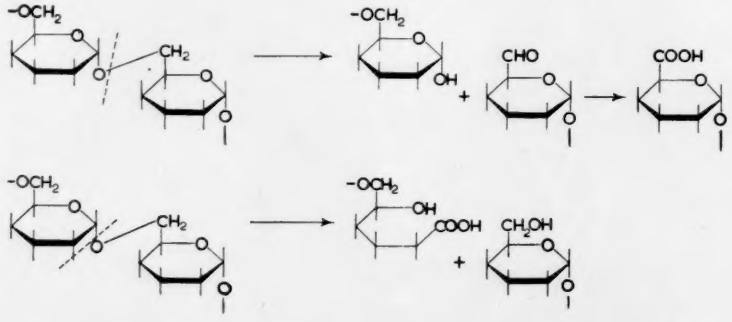



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TABLE 3. Irradiation of dextran in aqueous solution (0·15%)

| Compound | Specific activity of sample ($\mu$c/mmole of carbon) | Carrier (mmole) | Amount (mg) |
|---|---|---|---|
| Glucose (a) penta-O-acetate | $3{\cdot}5 \times 10^{-2}$ | 0·59 | 7·9 |
| (b) osazone | $1{\cdot}7 \times 10^{-1}$ | 0·21 | 7·1 |
| Erythrose | $7{\cdot}9 \times 10^{-1}$ | 0·004 | 0·45 |
| Glyoxal | $2{\cdot}7 \times 10^{-2}$ | 0·50 | 0·82 |
| Gluconic acid | $1{\cdot}2 \times 10^{-2}$ | 0·52 | 5·10 |
| Formaldehyde | $8{\cdot}3 \times 10^{-3}$ | 0·113 | 0·03 |
| Glucuronic acid | $1{\cdot}3 \times 10^{-2}$ | 0·41 | 4·5 |
| Dihydroxyacetone | $1{\cdot}08 \times 10^{-2}$ | 1·0 | 2·0 |

Glyoxal and erythrose, which are present in smaller amounts, are doubtless secondary products from glucose. Thus random attack along the glucose chain in dextran, as indicated in the formulae, would lead to all the main products observed. There are several other similarities between irradiated solutions of glucose and dextran.[17] These doubtless arise from the fact that glucose-containing polysaccharides suffer hydrolysis to glucose. The behaviour of glucose-containing polysaccharides[17] becomes intelligible on this basis and is in harmony with the observations of BOURNE *et al.*[22] on irradiation of aqueous amylose.

## 3. OXIDATION OF D-SORBITOL IN AQUEOUS SOLUTION

In a preliminary publication it was noted that irradiation of D-mannitol solutions leads to the formation of D-mannose.[26] Paper chromatographic examination of irradiated D-sorbitol solutions revealed the presence of glucose, gulose, arabinose and xylose in addition to unchanged D-sorbitol. Further confirmation was furnished by isotope dilution and this method also identified the particular optical isomer present. These were D-glucose, L-gulose, D-xylose and L-arabinose.

For irradiated D-sorbitol solutions, measurement of the yield of products with energy input is difficult because it is not possible to effect complete separation of the constituents present in the irradiated solution on paper chromatograms. In butan-1-ol-acetic acid–water, D-sorbitol moves at the same rate as glucose. In ethyl acetate–acetic acid–aqueous boric acid, D-sorbitol moves much faster than the products of irradiation, but in this solvent glucose and arabinose cannot be separated. By measuring the variation in radioactivity of the separated and combined spots with energy input in each solvent, however, yield/dose

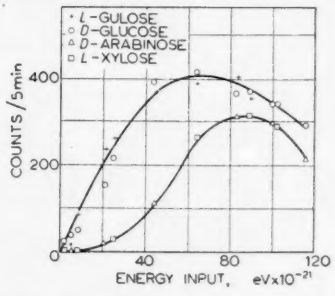



FIG. 3. Rate of formation of D-glucose, L-glucose, D-arabinose and L-xylose with energy input. Rate of energy input $1{\cdot}56 \times 10^{17}$ eV/min/ml.

curves for the individual products were obtained (Fig. 3). From these figures it is clear that the two hexoses and two pentoses are formed at identical rates. Therefore, during isotope dilution measurements only one hexose, D-glucose, and one pentose, L-xylose were quantitatively estimated. The results are shown in Table 4. Formaldehyde and glucuronic acid were also estimated.

*Mechanism of attack at primary alcohol group*

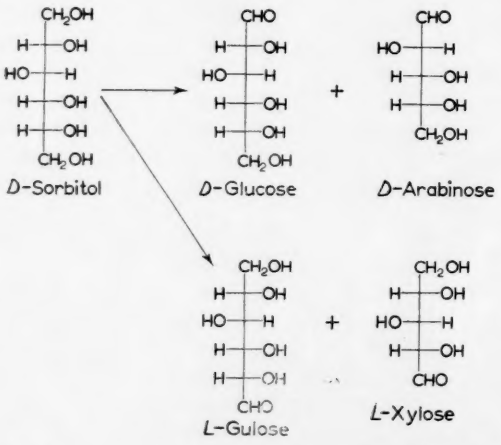
The behaviour of D-sorbitol on irradiation in aqueous solution brings into prominence

TABLE 4. Irradiation of aqueous solutions of D-sorbitol

| | |
|---|---|
| Initial D-sorbitol | 8·2 mmole |
| Energy input | $9{\cdot}3 \times 10^{22}$ eV |
| Specific activity | 3·8 $\mu$c/mmole |

| Compound | D-Sorbitol | D-Glucose | L-Xylose | Glucuronic acid | Formaldehyde |
|---|---|---|---|---|---|
| Carrier (mmole) | 0·48 | 0·54 | 0·36 | 0·54 | 0·113 |
| Specific activity ($\mu$c/mmole) | 1·03 | 0·53 | 0·26 | 0·065 | 0·335 |
| Yield (mmole) | 2·69 | 1·31 | 0·48 | 0·14 | 0·63 |

the greater vulnerability to free radical attack of the $-CH_2OH$ group in comparison with the $>CH-OH$ group. The main products are two hexoses and two pentoses formed by attack at the two extremities of the molecule:




While it was not possible to distinguish between D- and L-isomers by paper chromatography, the isotope dilution method established unequivocally that the pentoses were L-xylose and D-arabinose. No L-gulose was available for isotope dilution, but it may be inferred on the basis of the structural possibilities that the hexoses are D-glucose and L-gulose. According to the yield/dose curves, the hexoses are the primary products, and it appears that the pentoses arise from secondary attack on the hexoses as was described for D-glucose. The fact that the yields of the two hexoses are of the same order indicates that the two primary $CH_2OH$ groups are subjected to comparable attack by the radicals formed during primary radiolysis of water. A likely mechanism is as follows:

$$H_2O \rightarrow .H + .OH$$

$$.H + O_2 \rightarrow .HO_2$$

$$RCH_2OH + .OH \rightarrow \dot{R}CHOH + H_2O$$

$$RCHOH + O_2 \rightarrow RCHOH \quad (O_2 \text{ as } \dot{O}_2)$$

$$RCHOH + .HO_2 \rightarrow RCHO + H_2O_2 + O_2 \quad (O_2 \text{ as } \dot{O}_2)$$

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# Chemical Changes in Stored Foods when Microbial Spoilage is not Limiting

C. H. LEA

Low Temperature Research Station, Cambridge

The application of ionizing radiations to foods normally subject to rapid microbial spoilage can raise new problems, both by giving time for slower enzymic or chemical changes to assume significance and by accelerating some of the known processes of chemical deterioration. These factors are considered with particular reference to non-enzymic browning and lipid oxidations.

## INTRODUCTION

In the earlier papers of this meeting we have been discussing the production of ionizing radiations, and the physical and chemical effects which they produce in substances which are major constituents of foods. In later papers we shall be going on to consider the use to which the irradiation process can be put in destroying or inhibiting the multiplication of micro-organisms or other pests which infest food.

But first, I think it would be advisable to spend a little time in considering some of the changes which can occur in stored foods *other* than those brought about through the agency of micro-organisms: changes that are most likely to assume importance when the food, either through its composition or because of the conditions under which it is processed or stored, is relatively resistant to microbial attack.

Chemically, most foods are complex mixtures of reactive substances. Once the directive influence of the living cell has been disorganized, chemical interactions are liable to take place between the constituents, with resulting changes in appearance, texture, palatability or nutritive value which are often deleterious. Modern methods of food handling and processing limit and control such deteriorative processes in various ways. Enzymes which catalyse undesirable reactions may be inactivated by a "pasteurizing" or "blanching" process prior to storage. Moisture or oxygen may be removed or excluded by appropriate packing. Since most chemical reactions are slowed down in greater or less degree when the temperature of storage is lowered, many foods are preserved for long periods by chilling or freezing.

The application of ionizing radiations to foods normally subject to rapid microbial spoilage can therefore raise new problems simply by giving time for slower enzymic or chemical processes to assume significance. We also now know that radiation, while performing its desired function, often at the same time brings about a marked acceleration of some of the known processes of chemical deterioration, in addition, perhaps, to inducing new ones.

## ENZYMIC REACTIONS

In some cases, when bacteria have been inhibited by irradiation, reactions brought about by tissue enzymes may be found to take over as the factors primarily limiting storage life. Among many possible initiators of deleterious changes in foods are proteolytic or carbohydrate-splitting enzymes, causing breakdown of structure; lipolytic enzymes, which can cause rancidity in dairy products; lipoxidase, sometimes implicated



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in the development of "off" flavours in dried and frozen vegetables, and the polyphenol oxidases responsible for enzymic browning in fruits and vegetables. It is not my intention to spend any time in discussing this important subject, one aspect of which is being considered in a later paper from the Battelle Institute.

## NON-ENZYMIC BROWNING

Of the purely chemical changes to which stored foods are subject the rather complicated and ill-defined reactions grouped under the general title of "non-enzymic browning" are of particularly wide occurrence and interest. Browning reactions undoubtedly serve a useful purpose in contributing in major degree to the appetizing appearance and flavour of many of our cooked and heat processed foods. But in some heat processed foods and in most stored products where changes of this type occur the results are not appreciated and we do our best to avoid them.

*Symptoms of non-enzymic browning*

Deleterious changes associated with reactions of the browning type vary considerably from one food to another and may include any of the following: development of a brown discoloration ranging from a pale cream or biscuit shade to almost black; production of stale, caramelized, bitter or otherwise unpleasant odours or flavours; loss of solubility of the protein leading to deterioration in texture and to a failure of dried foods to reconstitute properly with water; a reduction of pH and the production of carbon dioxide and water; enhanced reducing properties leading to interference with the estimation of ascorbic acid with 2:6-dichlorophenolindophenol and of reducing sugars by the cuprimetric and ferricyanide methods; an increased tendency to froth or foam; the development of the property of fluorescing in ultra-violet light; and finally, a loss of nutritive value of the protein resulting from a reduced availability of certain of the essential amino acids, and a destruction of ascorbic acid when present.

*Conditions under which deteriorative browning develops*

The reactions responsible for browning proceed most rapidly when the reactants, which are often only minor constituents of the food, are present in concentrated aqueous solution. For this reason, browning is more prevalent in concentrated or dried than in fresh foods. A certain amount of moisture, however, is essential for most browning reactions and drying to a sufficiently low moisture content is a common method of control: on the other hand, a dried product allowed to equilibrate with a moist atmosphere offers nearly optimal conditions for deterioration of this type. The temperature coefficients of the chemical reactions concerned are high, and trouble may, therefore, be experienced in the tropics with materials which store reasonably satisfactorily in temperate climates; conversely, browning is rarely troublesome under chilled or frozen conditions of storage. Atmospheric oxygen is usually not essential for discoloration and may even decrease the extent of browning: in other cases browning is greatly accentuated in the presence of oxygen and other consequences of the reaction such as the nature of the "off" flavour produced and the amount of carbon dioxide evolved can be markedly affected.

*Nature of the constituents of foods concerned in browning*

Most of the carbohydrate constituents of foods will form brown caramelization products by a complicated process of dehydration and polymerization if heated sufficiently strongly. Reducing sugars such as arabinose, xylose, glucose and fructose decompose more rapidly than sucrose and starch, and glucuronic, galacturonic, ascorbic and reductic acids more easily still. These reactions can be accelerated by alkali or acid, and particularly by amino compounds, and then proceed extensively at much lower temperatures. In some foods, such as molasses, honey or maple syrup the amount of non-carbohydrate

materials involved may be so small that the browning reaction appears to be little more than a catalysed caramelization of sugar. In fruits and vegetables, organic acids can take part in complicated interactions with reducing sugars and amino acids; ascorbic acid when present in quantity is a potent precursor (via dehydroascorbic acid) of browning. In protein-rich foods such as fish and egg-white, comparatively minor proportions of sugars are able to produce serious changes in the proteins and amino acids which constitute the major part of the food.

In addition to proteins and amino acids, other nitrogenous food constituents such as the B-vitamins, thiamine, nicotinamide and *p*-aminobenzoic acid, can also undergo reactions of the browning type with reducing sugar, and this is probably one of the routes by which losses occur during the processing and storage of preserved foods.

Finally, in dried egg yolk, traces of glucose are known to react with the lipid amino groups of kephalin, with a resulting serious deterioration in palatability and colour.

*The mechanism of browning*

The mechanism by which amino compounds, particularly amino acids or proteins, accelerate the browning of reducing sugars is now fairly well understood, at least in so far as the earlier reactions in the sequence are concerned. Amino acids have been shown to react with aldoses to produce *N*-glycosides, which then isomerize according to the so-called Amadori rearrangement to give *N*-substituted 1-amino-1-deoxy ketoses. Many of these intermediate reaction products have been prepared from various amino acids and sugars. They are still colourless substances, from which, however, the sugar cannot be recovered intact by hydrolysis. On heating or on storage they readily undergo dehydration or fission to give reactive reductones or carbonylic fragments: these can then polymerize or react further with amino compounds to produce dark polymers and the nitrogen-containing co-polymers termed "melanoidins".

Hodge,[1] in a useful review of non-enzymic browning, has set out diagrammatically the inter-relationships between several of the reactions believed to be concerned in the production of melanoidins. In a very recent note Anet[2] has produced evidence that the Amadori-rearranged product from glucose and glycine (which we can call "fructose-glycine") is able to react with a second molecule of glucose, to give a "difructose glycine". This latter substance readily decomposes back again to fructose glycine, liberating the second sugar molecule in the form of reactive, unsaturated carbonylic fragments which readily undergo aldol condensation to brown polymers. By this or similar mechanisms the known catalytic effect of amino compounds in bringing about the decomposition of many equivalents of reducing sugar can be explained.

Inevitably, in so complicated a situation many uncertainties must exist as to the precise nature of the chemical processes involved, but the emphasis in most of the schemes proposed is on the formation of reactive carbonylic compounds as labile intermediates on the way to dark coloured polymers. Sulphur dioxide, which is the only widely used chemical inhibitor of browning, apparently does not interfere with the early, colourless stages of the amino–sugar reaction, but probably blocks subsequent browning in the various systems by inactivating reactive carbonylic substances which function as intermediates in the degradation of the sugars.

*The browning of irradiated foods*

In addition to providing more time for the natural constituents of foods to undergo possible browning reactions, irradiation can directly increase the tendency of foods to brown by producing in them compounds capable of undergoing this kind of change. Irradiation of individual carbohydrates and amino acids, and of various other food constituents, has been shown to give rise to labile carbonylic compounds, which can either undergo browning reactions themselves, or react with amino compounds already present in the food or produced also by the irradiation.

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TABLE 1. Influence of irradiation dosage and method of extraction on the carbonyl content* of ground beef and of pork fat[3]

| Dose (rads × $10^6$) | Ground beef | | Pork fat | |
|---|---|---|---|---|
| | Acid–salt extract | Benzene extract | Acid–salt extract | Benzene extract |
| 0 | 1·44 | 0·18 | 0·28 | 0·30 |
| 2 | 6·47 | 0·24 | 0·43 | 0·79 |
| 4 | 10·35 | 0·25 | 0·73 | 0·91 |
| 6 | 13·23 | 0·34 | 1·05 | 0·92 |
| 8 | 14·95 | 0·29 | 1·15 | 1·00 |
| 10 | 17·25 | 0·30 | 1·12 | 0·99 |

* as M × $10^{-5}$/g meat.

Foods which have been irradiated may, therefore, show an increased tendency to darken on cooking. We have repeatedly noticed that irradiated meats, particularly pork and chicken, tend to brown more than unirradiated controls under standard roasting conditions. SCHWEIGERT and his collaborators at the American Meat Institute[3] have measured the production of extractable carbonylic substances in irradiated meat, and find it to be much greater from the muscle than from the fat (Table 1). While these substances may not themselves have much direct effect on the odour or flavour of the meat it is known that some of them can react with odorous sulphydryl compounds or amines produced by the irradiation, and thus perhaps exert an indirect influence on palatability.

An increased tendency of meat to brown on cooking may not be objectionable, but the same property in milk certainly is. Milk can normally be boiled for quite long periods without darkening although it does, of course, brown perceptibly when sterilized in an autoclave, particularly when concentrated. But after irradiation it darkens much more easily. PROCTOR and his associates at MIT[4] have shown that skim milk, which had been irradiated with electrons at a dose level of 2 Mrads, developed a brown colour within a week when stored even at 40°F. When the tendency of whole milk to brown was tested by boiling for 20 min an effect of the radiation could be detected at as low as 50,000 or 100,000 rads, and the intensity of the colour increased linearly up to 2 Mrads. We have observed the same thing with irradiated whole milk powder (Table 4). Two distinct effects of the radiation are superimposed here, bleaching of the pigments occurring as well as browning.

The American workers showed that irradiated lactose solutions also darken on heating, and that the effect is enhanced when casein is also present. The isolation of crystalline 2:4-dinitrophenylhydrazones showed that carbonylic degradation products had been produced, and colorimetric estimation indicated the production of about 2 $\mu$M per ml of these substances for a dose of 2 Mrads (from 111 $\mu$M lactose/ml). These reactive carbonylic substances were obviously responsible for the increased susceptibility to browning, since browning was prevented when they were extracted from the solution, again by means of the 2:4-dinitrophenylhydrazine reagent. Continuous extraction with ethyl acetate of trace quantities of reactive carbonylic intermediate compounds, as they were produced, has previously been shown to prevent non-enzymic browning in apricot syrup which has not been irradiated. It would appear, therefore, that the general mechanisms of the ordinary and of the radiation-induced processes are probably similar. The presence of reductones (substances of rather similar structure and reducing properties to ascorbic acid) has also been demonstrated chemically in both systems, and further stresses their similarity. Amino acids as well as sugars have been shown by the MIT group[5] to give rise to reductones on irradiation, indicating that proteins can also act as browning precursors by this route.

## LIPID OXIDATIONS

A second important type of chemical change to be considered in relation to the preservation of foods by ionizing radiations is the reaction of lipids, directly or indirectly, with atmospheric oxygen.

Virtually all foods contain lipids, ranging

from the small amounts present in most fruits and vegetables to the major proportions present in most animal products. Deterioration as a result of lipid oxidation is by no means confined to foods of high fat content.

*Mechanism*

The great bulk of the constituent unsaturated fatty acids of food fats are of the non-conjugated, methylene-interrupted type which react with oxygen according to a free radical chain mechanism. *Initiation* of the reaction occurs by the removal of hydrogen from a reactive methylene group adjacent to a double bond, to give a resonance stabilized free radical which, in the presence of air, usually adds on oxygen to give hydroperoxide radicals. These, in turn, abstract hydrogen from further lipid molecules to *propagate* the reaction chain, being themselves converted to conjugated hydroperoxides. *Termination* of the chain will usually be by reaction of the chain propagating radicals, either together to give non-radical products (polymers), or with an antioxidant to give a radical insufficiently reactive to carry on the chain.

Naturally, oxidation of this type occurs most readily when a methylene group is doubly activated by being sandwiched between two double bonds, and the reaction rates for dienoic and more highly unsaturated acids containing the pentadiene grouping is very much greater than for monoethenoid acids. Saturated fatty acids are much more resistant still, and are only peroxidized extensively under severe conditions.

*Oxidation products*

Peroxides are therefore the major initial products of the autoxidation of edible fats, and it is by the chemical determination of these peroxides that we most commonly estimate the degree of oxidation that has occurred. But fat hydroperoxides are labile substances which can react further, by fission, dehydration, polymerization or further oxidation, to give a variety of degradation products, including the volatile aldehydes, ketones and acids which are largely responsible for the objectionable odours and flavours. Hexanal, 2-octenal and 2:4-decadienal, which have been isolated from autoxidized cottonseed oil, could be derived, for example, by fission of the isomeric hydroperoxides formed by oxidative attack at the $C_{11}$ reactive methylene group of linoleic acid. But many alternative routes of decomposition are possible, particularly of the polyunsaturated fatty acids which are present in major or minor degree in many edible fats, and a wide variety of volatile carbonylic decomposition products have been isolated in recent work from various oxidized fats and foods. We have been determining flavour thresholds on some of these aldehydic decomposition products of autoxidizing fats and find them detectable at concentrations as low as one part in $10^8$ or $10^9$.[6]

TABLE 2. Effects of lipid oxidation

Unsaturated fat
+
$O_2$

| Peroxides | Coupled oxidations |
|---|---|
| | Cause destruction of |
| | (a) Vitamins A, C, E, etc. |
| | (b) Haemoglobin (discoloration of meat) |
| Aldehydes, short-chain acids, polymers etc. (Cause "off" odours and flavours; discoloration; destruction of "essential" fatty acids; possible toxicity; "Browning" reactions with protein) | (c) Carotenoids, chlorophyll, anthocyanins (discoloration and "off" flavours in vegetable products) |
| | (d) Flavour and aroma constituents |

It should be stressed that these odorous, volatile aldehydes are accompanied by very much larger quantities of non-volatile carbonylic substances, probably unsaturated glycerides containing keto groups. These, like the hydroperoxides, are not likely to contribute in major degree to "off" odour and flavour but are probably important as precursors of the volatile, odorous compounds.

But it is not only the oxidation of the fats themselves which causes deterioration; other constituents of foods are oxidizable too, and tend to be destroyed when the fat oxidizes (Table 2). Probably they are largely

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destroyed by reaction with the free radicals produced during the chain oxidation of the fats. Pigments, flavour constituents and vitamins are only minor components of food on a weight basis, but they are very important in determining appearance, palatability and nutritive value, and damage to them can easily make a food unacceptable. The oxidation of carotenoids, for example, can adversely affect palatability as well as colour.

*Factors which influence the oxidation of lipids*

A feature of the reaction of unsaturated fats with oxygen is the facility with which the rate and extent of the reaction can be influenced by factors other than the usual ones of concentration and temperature (Table 3).

TABLE 3. Factors which influence lipid oxidation

Composition (degree of unsaturation)

| *Acceleration by* | *Inhibition by* |
|---|---|
| High temperature | Refrigeration |
| Light (u.v. and blue) | Opaque or coloured wrappers or containers |
| Ionizing radiations | Exclusion of oxygen |
| Peroxides (including oxidized fats) | |
| Lipoxidase enzyme | "Blanching" or other heat treatment |
| Organic iron catalysts (haemoglobin etc.) | |
| Trace metal catalysts (Cu, Fe, Mn etc.) | Conservation of natural antioxidants |
| | Addition of permitted antioxidants or metal deactivators |

Heating sometimes markedly *increases* the stability of fatty foods towards oxidation, apparently owing to the formation of substances with antioxidant or metal deactivating properties, e.g. sulphydryl compounds or products of browning reactions.

The lipids of foods already contain natural antioxidants, among which the tocopherols (Vitamin E) are prominent. Seven homologous tocopherols are now known, of varying vitamin E and antioxidant activity. Because animals cannot synthesize tocopherols and have to obtain them from the feed, the tocopherol contents of animal fats are usually much lower than those of vegetable origin.

Sometimes the fat of a food can be usefully stabilized by the incorporation of antioxidant-active edible material such as spices, citric acid or cereal, or by smoking, (which deposits phenolic antioxidants, mainly derivatives of catechol and pyrogallol). Finally, many countries now permit the addition of chemical antioxidants in limited quantities to certain classes of foods, usually in the main to edible fats and manufactured foods containing them.

*Effects of irradiation on lipid oxidation*

Since the autoxidation of unsaturated fats is initiated and propagated by free radicals it is not surprising that ionizing radiation accelerates the normal oxidation process. Because of the high concentration of radicals produced by the radiation the numbers of chains started will be greater than in ordinary autoxidation, and their average length is likely to be shorter and the effectiveness of antioxidants in inhibiting the oxidation less.

Considerable evidence has been produced of a general similarity between the autoxidative and radiation-induced processes: SLOVER and DUGAN,[7] for example, find the same four isomeric hydroperoxides produced in approximately the same proportions by $\gamma$-irradiation of methyl oleate as by autoxidation. Discrepancies in the proportions of the volatile carbonylic products isolated by GADDIS and ELLIS[8] from pork fatty tissue which had been allowed to become rancid in cold storage, and those isolated by WITTING and SCHWEIGERT[9] from $\gamma$-irradiated lard may be due partly to differences in technique. It is known that differences in the proportions of reaction products can be caused by factors such as the amount of oxygen available, the temperature during oxidation or extraction of the products,* exposure to light or the presence of metallic catalysts, non-fatty materials etc. On the

* GADDIS and ELLIS pointed out that high temperatures such as used in cooking (e.g. 165°C) greatly increase the proportions of conjugated unsaturated carbonyls produced; WITTING and SCHWEIGERT removed their volatiles in steam at 150°C.

other hand, the great predominance of hexanal in GADDIS and ELLIS's results and of 2:4-undecadienal in those of WITTING and SCHWEIGERT might suggest that linoleic acid was being attacked mainly at carbon 11 in the former and at carbon 8 in the latter case.

Certainly the effects of irradiation appear to go beyond those of normal autoxidation in a number of respects. There would seem, for example, to be a less selective degradation of the more unsaturated constituents; even saturated fatty acids, which are resistant to autoxidation under ordinary storage conditions, show appreciable peroxidation under the influence of irradiation.[10] Moreover, changes resulting in the production of carbonylic compounds and "off" flavours have been observed on irradiation even with rigid exclusion of air, the necessary oxygen presumably being derived from the ester groups of the glyceride molecules.[10] Chemical interactions between, for example, unsaturated carbonylic and SH– or $NH_2$– containing irradiation products may, as already indicated, have important effects on the palatability of irradiated foods.[9]

I propose to conclude with a very brief reference to some recent experiments of our own in which comparatively low levels of irradiation have sufficed to induce appreciable oxidative changes in normally quite resistant foods.

*Chilled beef fat*

In collaboration with Macfarlane we irradiated external fatty tissue from forequarters of beef, and stored it at −1°C and controlled relative humidities. The experiments were continued for periods of up to 3 months, to simulate overseas transport of chilled beef. The results showed that doses of 50,000 or 100,000 rads produced a very appreciable retardation of microbial spoilage, as indicated by a delay in the development of "tainted" odour and flavour, by a retarded appearance of visible bacterial or mould growth and by a slower rate of increase in the free fatty acid content of the fat.

But symptoms of accelerated fat oxidation were also present, as indicated by the development in the exposed surface of a tallowy odour and flavour, which was easily perceptible in the raw or lightly cooked fat. The yellow carotenoid pigment tended to be noticeably bleached in the superficial layers of the irradiated tissue although, owing to the non-uniform distribution of the pigment, it was not always possible to follow bleaching quantitatively in the extracted fat. Peroxide accumulation was consistently more rapid in the irradiated than in the control samples.

The tasting panel did not object to the "tallowy" odour of the irradiated fats so much as to the "tainted" flavour of control samples which had been more severely attacked by micro-organisms. But the margin of safety, if any, is obviously small, and it is likely to be influenced by the degree of initial contamination and by subsequent storage conditions.

*Milk powder*

Data were obtained on chemical changes in irradiated roller-dried milk powder, not because there is any need for the irradiation preservation of this product, but because we wished to try out a simplified procedure for the direct determination of volatile carbonylic flavour substances in dried foods.[11] The effect of 50,000 rads on the appearance and palatability of the powder was only slight (Table 4). Roller-dried milk powder has, of course, a pronounced "cooked" odour and flavour which would probably mask flavours easily detectable in fresh milk. With 100,000 rads there was serious browning, fat oxidation and bleaching of the pigments. Peroxide and total carbonyl values determined on the extracted fat, and the volatile carbonyl values determined directly on the powder gave generally similar pictures of the effects of the radiation.

Powder irradiated with 100,000 rads also oxidized rapidly during a post irradiation storage period of 5 days, whereas roller powder normally resists oxidative spoilage for periods of the order of a year.

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TABLE 4. Effect of irradiation with $\gamma$-rays on the properties of roller-dried whole milk powder

| Dose (rads) | Appearance of the powder | Reconstituted in warm water | | |
|---|---|---|---|---|
| | | Odour and flavour | Appearance | Appearance after boiling 20 min |
| 0 | Yellowish white | Normal, "cooked" | Normal | Normal |
| 50,000 | Yellowish white | Very slight "off" flavour | Very slight bleaching | Very slight bleaching |
| 100,000 | Slight bleaching | Definitely tallowy | Obvious bleaching of the fat* | Just detectable buff shade |
| 1,000,000 | Very slightly buff shade instead of pure yellow | Extremely unpalatable | Chalky white and completely bleached | Slight but easily detectable buff colour |

* Most easily visible in the fat globules which separate on the surface on standing.

TABLE 5. Effect of irradiation and subsequent storage (5 days at 25°C) on the flavour and carbonyl content of roller dried milk powder

| Sample | Flavour score* | $\mu$M/g fat | | | |
|---|---|---|---|---|---|
| | | Total CO | | Volatile CO | |
| | | Unsaturated | Unsaturated + Saturated | Unsaturated | Unsaturated + Saturated |
| Control | 0 | 0·5 | 10·5 | 0·02 | 0·08 |
| Control stored | 0 | 1·4 | 8·6 | 0·03 | 0·11 |
| 50,000 rads (dose) | 0·5 | 1·9 | 9·3 | 0·03 | 0·11 |
| 50,000 stored | 2 | 2·9 | 8·9 | 0·06 | 0·13 |
| 100,000 rads (dose) | 2 | 4·2 | 14·7 | 0·10 | 0·28 |
| 100,000 stored | 3 | 11·1 | 27·2 | 0·59 | 1·11 |
| 1,000,000 rads (dose) | 4 | 62·3 | 86·1 | 1·25 | 2·48 |
| 1,000,000 stored | 4 | 47·8 | 133·4 | 1·45 | 5·39 |

* 0 = Same as control; 1 = Very slight off-flavour; 2 = Definite off-flavour; 3 = Unpleasant; 4 = Very unpleasant.

American work has shown that such oxidative effects in fats can often be greatly reduced by irradiation and subsequent storage in the absence of atmospheric oxygen, but this may not always be feasible.

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# Discussion

Professor DESCHREIDER (Belgium): In our Laboratory it has been found that the phospholipids from flour starch show an increased adsorption on the starch particles after the flour has been irradiated with $\gamma$-rays.

We have also found that brown aggregates are formed in the aqueous extract of irradiated flour. These aggregates contain the same amino acids as the brown pigment of the crusts of bread. This could explain the abnormal coloration of the crumbs of bread made from flour irradiated with $\gamma$-rays.

The polymerization or condensation of the proteins of meat mentioned by Dr. Schweigert has also been observed in our laboratory with the proteins of flour. It reveals itself with irradiated flour by modifications to the distribution of the various proteins and soluble amino acids in different solvents.

Dr. PHILLIPS (U.K.): I wonder if Dr. Schwiegert can elaborate upon the comments which he made about the polymerization reactions between amino acids and reducing sugars during irradiation. Do these occur in the presence or absence of oxygen?

Dr. SCHWEIGERT (U.S.A.): I was referring to some unpublished studies by Dr. Wolfrom and his colleagues at Ohio State University. These workers observed the formation of polymeric components at doses of 10 to 20 Mrads. These products resemble those formed in the browning reaction of foods, and presumably the reaction involves the amino groups of glycine and the aldehyde group of the sugar. Several components have been observed in chromatographic separations made on the mixture that was irradiated. I think that the experiments were made in the presence of oxygen, but I cannot be certain on this point.

Dr. MOCQUOT (France): When he referred to the formation of large polymers as the result of irradiation of aqueous solutions of certain purified milk proteins, Dr. Schweigert mentioned that constituents different from those of the original protein appeared in the irradiated protein.

Is it a matter of changes in those constituents of the proteins that can be separated by electrophoresis? Has this been established by reproducible experiments, either by simple electrophoresis or by immuno-electrophoresis?

Dr. SCHWEIGERT (U.S.A.): Relatively little work has been done, other than the physical measurements referred to, on the intact aggregated material formed during irradiation. We know disulphide bonds are formed in proteins containing sulphydryl groups and that the sedimentation constant of irradiated myoglobin is altered. The specific types of bonds in the polymers formed have not been fully elucidated. It is clear from the electrophoretic work that there is a net change in the ratio of the acidic and basic groups present in the protein material.

Dr. SCHRAM (Belgium): What effect do the changes in proteins observed on irradiation have on the growth of micro-organisms?

Dr. SCHWEIGERT (U.S.A.): I do not know.

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# Fundamental Radiobiology

H. LASER

Medical Research Council, Molteno Institute, Cambridge

Food sterilization by ionizing radiations is much dependent on our knowledge of the basic effects of these radiations on the one hand on micro-organisms, the growth of which it is intended to suppress, and, on the other hand, on many components of the biological material itself, as for example, specific proteins like a host of enzyme systems, or on myoglobin, or on fats, etc. A few examples pertaining to each class are given, demonstrating how changing conditions prior to or during irradiation, including variations of oxygen tension, affect radiosensitivity.

A discussion of fundamentals in radiobiology, in contradistinction to and complemented by the important field of radiation chemistry, requires the study of:

(i) the functional and biochemical state of the biological entity, which is affected by irradiation, *at the moment of irradiation*;
(ii) the biochemical changes which follow in the course of irradiation;
(iii) the alterations or loss of cellular functions, which these changes cause;
(iv) the possible means by which unwanted changes might be attenuated and desired changes facilitated.

Some of these points will be briefly discussed by means of examples dealing with X-radiation of bacteria, enzymes and some substances of biological importance.

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## BACTERIA

The most important effect of radiation on bacteria is, obviously, their "killing" effect, in relation to which the following points are now generally agreed, namely, the survival curve for a given organism in pure culture is within wide limits independent of the concentration of organisms, and secondly, the viable count decreases exponentially with increasing dose. It must, however, be remembered that these facts are not *a priori* synonymous with cell death from the biochemical point of view, but consequences which become apparent as loss of certain functions with time. Thus, for example, the respiration of *E. coli* B in presence of glucose immediately after both aerobic and anaerobic X-radiation, which decreases the viable count by >90 per cent, proceeds for several hours at the same rate as that of unirradiated controls.[1] Only then does the respiratory rate begin to fall off to a lower level. In our own experiments it was found, at doses from 16 to 64 krads, to drop to a level varying between 15 per cent and 80 per cent respectively of that of the control. The inhibition of the respiratory rate ultimately effected was about 2–3 times greater after irradiation in air than in $N_2$. Similarly, other functions, such as aerobic or anaerobic fermentation or glycolysis remain unaffected for some time after irradiation and cell division is not as a rule immediately inhibited, one division frequently proceeding at the normal rate. Thus the radiation seems to cause the loss of different functions at relatively different rates with time, the most significant of these being stoppage of cell division, presumably as a sequence to radiation induced interference with DNA synthesis,[2] which itself may not be the primary effect of radiation, but the result of blockage in some other event in the development of the cell.[3]

*Modification of radio-sensitivity*

The same dose applied to the same type of bacteria does not always produce the same effect. This was most convincingly shown in the important publication by HOLLAENDER *et al.*[4] who not only demonstrated the existence of an "$O_2$-effect" in *E. coli*, but found that the sensitivity varied greatly according to whether the bacteria prior to irradiation had been grown under aerobic or anaerobic conditions, the sensitivity being greatly reduced in the latter case. There is no doubt that the enzymic make-up of the aerobically and anaerobically grown strains is different—the latter probably having a depleted complement of cytochromes—and that this difference is likely to be responsible for the varying response to radiation. That enzymic composition and functional state within the cell contribute to determining radio-sensitivity was furthermore shown[5] when the $O_2$-effect, that is greater inhibition of growth after aerobic than after anaerobic irradiation, was abolished in spores of *B. subtilis* by freeze-drying the spores prior to irradiation, or in low-nitrogen yeast, that is yeast which had been aerated for prolonged periods in presence of glucose without an added nitrogen source, and finally in *Sarcina lutea*, when irradiated in presence of certain respiratory inhibitors, as e.g. KCN, CO and hydroxylamine. Lastly it should be mentioned that irradiation of *Exherichia coli* B in $D_2O$ in presence of air, increased their sensitivity about three-fold.[6]

*Enzymes*

It is the generally accepted view that ionizing radiations affect enzymes through the formation of products resulting from the ionization of water, that is through an indirect effect. This follows from the work of DALE[7] on carboxypeptidase, of SUTTON[8,9] on catalase, and of YOST *et al.*[10] on tyrosinase. Destruction of the enzyme by radiation has usually been measured by determining the decrease in rate of substrate utilization, which resulted from the irradiation. This tells us little about the actual point of attack in the enzyme molecule. However, SUTTON,[9] in his most valuable contribution, has shown for catalase, that its own substrate, $H_2O_2$, and cyanide, both of which combine with ferric iron, confer a high degree of protection, when present during the irradiation. This evidence is conclusive that the major inactivating effect of X-rays on catalase in solution is localized on the iron centres, while the protein portion may be damaged by radiation without loss of catalytic activity.

However, protection of an enzyme by its specific substrate is not the general rule. Thus DALE found that carboxypeptidase and D-amino-acid oxidase[11] could be largely protected not only by their specific substrates but by a variety of structurally unrelated substances, which may have acted as scavengers for radicals. On the other hand, LASER[5] found that the flavin enzyme notation (glucose oxidase) was fairly radiation resistant, when given 38,000 rads in air, but became very sensitive, i.e. 60 per cent destroyed, when irradiated in presence of its specific substrate, glucose. This effect was specific for glucose, since it could not be brought about by a variety of sugars which had been tried.

From the point of food sterilization, bacteria in meat are certainly well supplied with their specific substrates. It must therefore remain a matter of speculation, whether inactivation of intracellular enzymes is thereby speeded up or suppressed, quite independently of the question of oxygen supply during irradiation. It may also be noted, that under certain conditions, namely those requiring induced enzyme formation by the bacteria in order to metabolize a new substrate, viable cells remaining after irradiation may do better and grow at a faster rate than unirradiated controls.[12]

*Substances of biological importance*

Experiments with irradiation by X-rays of cytochrome-*c* and haemoglobin will be briefly reported, the latter being of special interest in view of its great similarity with the myoglobin of meat. The primary change produced in haemoglobin solutions was found to depend on the initial state of the material, whether oxidized or reduced, and to be

independent of the presence of oxygen.[5] Thus haemoglobin becomes oxidized and methaemoglobin becomes reduced. The oxidation of oxyhaemoglobin in air and of haemoglobin in nitrogen is of about the same order, while reduction of methaemoglobin is greater in nitrogen than in air. Oxidation or reduction, however, is in neither case directly proportional to the X-ray dose, because of the occurrence of secondary reactions. Thus, for example, methaemoglobin, which becomes reduced during irradiation in nitrogen (presumably by hydrogen atoms), is re-oxidized (presumably by hydroxyl radicals). Moreover, it is not re-oxidized to methaemoglobin, but to a green pigment showing a strong band at 615 m$\mu$. The rate of this secondary oxidation is greater in nitrogen than in air, and in both cases it is the greater the lower the concentration of haemoglobin. Similar results have been obtained by GINGER *et al.*[13] with myoglobin, including both oxidation and reduction and formation of the green compound, absorbing light at 610–620 m$\mu$.

*Cytochrome*-c. Like haemoglobin, reduced cytochrome becomes oxidized both on irradiation in air and in nitrogen, and ferricytochrome becomes reduced on irradiation in nitrogen. Unlike haemoglobin, on the other hand, ferricytochrome does not appear to be affected by irradiation in air at doses up to 35,000 rads. There is, furthermore, no formation of a green pigment from either ferri- or ferrocytochrome during or after irradiation in air, nor during irradiation in nitrogen. However, cytochrome irradiated in nitrogen turns rapidly green on admission of oxygen after irradiation. This occurs irrespective of whether the sample had been partially oxidized or reduced during irradiation, and is accompanied by the disappearance of the band of ferrocytochrome at 550 m$\mu$ and the appearance of a new band at about 600 m$\mu$. The catalytic activity of cytochrome after a dose of 38,000 rads in air was decreased by only 15 per cent, while the same dose applied in $N_2$ resulted in an activity loss of 35 per cent, due to that fraction of cytochrome which had been irreversibly oxidized to the green pigment by molecular oxygen.

Lastly, experiments on irradiation of riboflavin may be briefly mentioned. There too, both oxidation and reduction on irradiation in air as well as in $N_2$ are encountered, which again are not directly proportional to the X-ray dose applied, because of the occurrence of secondary reactions.

The question of the "$O_2$-effect", that is in general the greater efficiency of a given dose of ionizing radiation when applied in oxygen than under anaerobic conditions, has only been mentioned in passing. In the opinion of the writer and in view of the clear evidence that the state of the cell *at the moment of irradiation* determines, at least in part, its radiation response, there seems to be no pressing need for the formulation of theories which postulate involvement of ionization of special vital target structures, which are prevented from returning to normal function by the interference of oxygen[14] or the formation of a carbon radical–oxygen complex in molecules that are essential to the cell for successful reproduction.[15]

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# Radiation Microbiology as Applied to Foods

T. HORNE* and B. A. BRIDGES

Technological Irradiation Group, Isotope Division,
Wantage Radiation Laboratory, Wantage, Berks

There is no such thing as a predictable "inactivation dose" which will just kill off with 100 per cent certainty, all the micro-organisms in a given sample. The inactivation of most species of micro-organisms by ionizing radiation is approximately an exponential process and doses suitable for sterilizing foodstuffs must be calculated with this fact in mind. It is shown that, assuming a standard of sterility of $10^{-8}$ organisms/g, a safe sterilizing dose will be of the order of 3–5 Mrads according to the initial concentration of micro-organisms. The effect during irradiation of certain environmental factors such as pH, freezing, heating, additives etc. is discussed in relation to food processing and stress is laid on the fact that we still know comparatively little about post-irradiation effects. Because of the undesirable quality changes in most foods after 3–5 Mrads of ionizing radiation, it is suggested that low dose and combination procedures might prove more practicable. There is still need for many more fundamental microbiological studies before progress can be made on a large scale.

## INACTIVATION OF MICRO-ORGANISMS

In pure culture most micro-organisms are inactivated exponentially, that is to say the semi-logarithmic plot of fractional survival against dose is a straight line. Two other types of curve have been described[1] but they are not commonly found in practice and will not concern us here. In mixed culture, each type of micro-organism will be inactivated independently of the presence of other types and this fact is of some importance when dealing with foods. Since different micro-organisms have different sensitivities to radiation the inactivation curve for such a mixed culture will not be a straight line but will tend to be concave upwards. Most of the early work on radiation inactivation was confined to determining the minimum dose with which no survivors could be detected in a small number of heavily contaminated samples. The results consequently are of limited value, being only semi-quantitative, but may be used to illustrate the different sensitivities of various species. Among the most sensitive are *Pseudomonas sp.*, frequently found causing meat spoilage; these may be killed off in pure culture by as little as 30,000 rads. Most vegetative bacteria however need between 50,000 and 500,000 rads for inactivation, but yeasts, moulds and many bacterial spores may need up to 1 Mrad. By far the most frequently encountered radiation-resistant organisms are bacterial spores. Those of *Bacillus subtilis*, *Clostridium sporogenes*, and *Clostridium botulinum*, which may require over 2 Mrads, are frequently used as representative of this group. There are however, certain resistant micrococci even more resistant than these spores[2] which, although rare, could prove troublesome with certain foods.

### *Sterilizing dose*

In the preceding section, we have discussed some of the doses required to inactivate various micro-organisms. These doses were determined using a few heavily contaminated samples. Commercial sterility, however, is usually determined with many samples having a comparatively low initial count of

* Present address: Princeton Division, Curtiss Wright Corporation, Princeton, New Jersey, U.S.A.

micro-organisms. It has been suggested[8] that $10^{-8}$ organisms per g is a common standard of sterility with heat processed foods, and there is no reason why this standard should be relaxed for radiation processed foods. This means that the inactivation factor will lie between $10^8$ and about $10^{14}$ according to the initial degree of contamination of the food. With normal techniques one cannot investigate inactivation factors much greater than $10^7$, and so with pure cultures of bacteria one is reduced to extrapolating the exponential curve to the values required. There is no evidence that any great errors are introduced by this step when dealing with pure cultures of micro-organisms, although certain viruses may provide exceptions.[3]

With mixed cultures such as are found in foods, the semi-logarithmic survival curve is not a straight line and therefore cannot be extrapolated. PROCTOR and GOLDBLITH[4] have suggested that the commercial sterilizing dose may be obtained by extrapolating the survival curve for the most resistant organism found in the food, starting at the most probable concentration of this organism. This is a fairly good approximation since inactivation of a given species is independent of the presence of other species. However, if a less resistant organism is present in excessively large numbers, these may survive after the smaller numbers of resistant organisms have been inactivated, although this is unlikely to occur in practice. *Clostridium botulinum* is a suitable organism for this calculation since it is very nearly the most resistant spore-former and is believed to be the most resistant pathogen found in food. It is taken here as representative of the radiation resistant spore-forming organisms found in food, most of which are, however, non-pathogenic, and of which *Cl. botulinum* spores will form but a small proportion. In Fig. 1 the survival curve (taken from some data of MORGAN and REED[5]) has been extrapolated down to $10^{-8}$ organisms per g. It can be seen that for a food which is only lightly contaminated with resistant spores, say between 1 and 10 per g, the calculated sterilizing dose is about 3 Mrads. For a food containing $10^3$ resistant spores per g, the sterilizing dose would be 4 Mrads. It should be stressed that these figures are only designed to give a rough idea of the order of the dose of radiation which would be needed. Where radiation-resistant micrococci are likely to occur, as in certain meat products[2,6] the dose may have to be higher still. Foods will have to be investigated individually since some of them contain substances capable of altering the sensitivity of micro-organisms with a consequent change in the sterilizing dose.

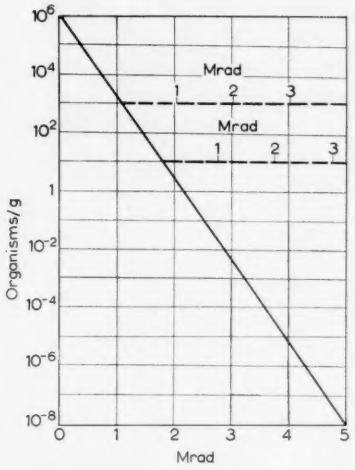


FIG. 1. Sterilizing doses for resistant spore-formers (*Cl. Botulinum*, extrapolated from data[5] of MORGAN and REED).

As with other forms of processing, a great reduction in sterilizing dose could be obtained if a reliable method could be devised of inducing germination of bacterial spores beforehand. This could be done either with a short incubation or by chemical means. KAN *et al.*[7] have published some promising results in which pre-irradiation germination was effected by alanine or adenosine, and in which the sensitization to radiation was greater than to heat, but there is no method available which will give 100 per cent germination every time.

*Low dose procedures*

Two main types of process are relevant here. First there is the complete eradication of a sensitive pathogen, an example being

the destruction of *Salmonellae* in whole egg pulp. The dose necessary may be calculated by extrapolation as before. Secondly there is the procedure of irradiation with 50,000–100,000 rads aiming, not at sterilization, but at greatly lowering the microbial count and thereby extending the shelf life of the product. This process is frequently termed "radio-pasteurization". The main snag with this procedure is that the residual microbial flora is different from the normal, and spoilage, when it occurs, may be of a different type from normal, perhaps a type not easily recognizable by the consumer. It has been estimated[8] that in general, a dose of 0·1 Mrad would reduce the microbial population of a food product by a factor of $10^2$ or $10^3$, and that this might be expected to give an increased storage life with many foods, especially at refrigeration temperatures.

Low or moderate doses may also be used in conjunction with other factors such as acid conditions, low temperature storage and heat treatments.

*Factors affecting radiation inactivation*

Besides the initial count of micro-organisms, sterilization and pasteurization doses are dependent on environmental factors before, during and after irradiation.

When it is realized that pasteurization depends for its success on a large reduction in the number of vegetative cells, and sterilization on complete elimination of bacterial spores, it will be seen that environmental factors may affect the two processes in different ways. Bacterial spores are frequently much less affected by changes in their environment during radiation than are vegetative cells. The effect of a wide range of environmental factors on the sensitivity of micro-organisms to radiation has been recently reviewed by Bridges and Horne.[9] In this paper we would like briefly to discuss six of these factors, which are relevant to radiation preservation; pH, freezing, heating, age of cells, composition of the gas phase, and chemical additives. The pH of the medium has no large effect on the sensitivity of bacteria to radiation although both spores and vegetative organisms have been shown to be more resistant at neutral pH.[12,20] It is possible that radiation might be used to kill off moulds and yeasts under conditions where a low pH prevents the growth of other micro-organisms, but this is a combination process rather than an environmental modification of radiation sterilization.

Freezing might be expected to protect against ionizing radiations by limiting the mobility of free radicals formed in the water. This has been found to be so with vegetative organisms[10–13] but bacterial spores have, in several reports,[12,14,15] been shown to be more sensitive in the frozen state. This fact must be considered in food technology because it may be necessary to irradiate certain foods in the frozen state in order to minimize damage to the flavour of the food.[8]

The use of heat either before, during or after irradiation can increase the effect of radiation on vegetative organisms and spore-formers, although the mechanisms may be different since the effect of the heat appears to be greatest after irradiation for spores[5,16,17] but before or during irradiation for vegetative cells.[13,18,19] Pre-irradiation heating might also be expected to induce germination of spores into the more sensitive vegetative forms, but this does not appear to have been sufficiently investigated as yet.

There would seem to be some agreement that bacteria are at their most sensitive in the logarithmic phase of growth.[21,22] Whether this fact can be used in radiation sterilization, for example by giving a short (say 2 hr) incubation period prior to irradiation, remains to be seen.

The composition of the gas phase has a very marked effect on microbial radiosensitivity. Oxygen has the most marked effect, increasing the sensitivity by a factor of three.[21] This effect would doubtless have been used in radiation preservation technology were it not for the fact that damage to the food itself is also increased by a comparable factor. For this reason one might be restricted to packing *in vacuo* or in nitrogen, irrespective of bacterial effect. Packing *in vacuo* is, however, advantageous

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in controlling microbial growth after irradiation, as INGRAM describes.[26]

The presence of chemicals during irradiation may have a protective effect, an enhancing effect, or may have no effect. Protective chemicals are frequently present in the foods themselves[24,8] and this should be taken into account when calculating sterilizing doses. It has also been suggested that certain chemicals might be added to minimize the production of off-odours and flavours in foods during irradiation,[8] but such chemicals would tend to protect micro-organisms as well.

The use of chemicals to enhance radiation damage to micro-organisms is at present of academic interest only owing to the extremely small number of such agents reported, and their low degree of sensitization achieved.[7,25]

It is but a short step to the use of environmental factors as processes in their own right in combination with radiation, and these are discussed by INGRAM.[26]

One point which is not often mentioned and which we feel should not be overlooked concerns the state of micro-organisms surviving a radiation process. It is a well known fact that those which survive a commercial heat process are frequently in a dormant state and may remain in such a state for long periods of time, especially in canned foods, although they are viable when transferred to another growth medium. It is not known whether bacteria surviving irradiation may be kept in a similar dormant state. The work of STAPLETON[21] and others has shown that certain bacteria may recover from the effects of radiation if they are incubated at sub-optimal temperatures or in the presence of certain metabolites, and it would seem desirable that further work be done on this topic.

## CONCLUSION

We do not feel that we would be justified in drawing a very optimistic conclusion with regard to radiation sterilization. Radiation damage is fairly extensive at the doses which are necessary for commercial sterilization and it is the responsibility of the microbiologist to investigate the effect of environmental factors and to devise conditions where the lethal effect of radiation might be expected to be at its greatest. Such steps as have been made in this direction have been of little use, either because the effect on radiation killing is small, or because damage to the food is increased by a comparable amount. There is much more work to be done in this field, however, and time must be allowed for the completion of fundamental studies before any large steps forward can be made.

The use of low doses of radiation either as pasteurization procedures or in combination with other treatments is more promising, and other speakers will elaborate on these topics.

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# Combination Processes

M. INGRAM

Low Temperature Station for Research in Biochemistry and Biophysics,
Department of Scientific and Industrial Research, Cambridge

Combination processes are here regarded as those whose influence is exerted primarily during the period after irradiation. As such the following are considered: refrigeration, vacuum-packing, addition of antibiotics, curing and heating. The first three do not change the state of raw foods.

The value of refrigeration, besides the general slowing down of enzymic, chemical and microbiological changes, is that temperatures below 5°C prevent the development of food poisoning organisms: hence chilling seems necessary to supplement a process of radiation pasteurization which might allow them to survive. Vacuum-packing, and antibiotics, though they further prolong the storage life of irradiated foods, can also not be relied on to inhibit pathogens, so that refrigeration is desirable when using these processes too.

Some cured products resist high doses of radiation and can thus be sterilized; but others are more susceptible and tolerate only small pasteurizing doses, when there might apparently be a hazard from staphylococci.

The combination of heating and radiation, for sterilization, is interesting because irradiation might replace part of the often excessive heat treatments given to canned foods. Even short of sterilization, irradiation should confer a high degree of stability on cooked foods: though sporing anaerobes would be likely to survive, making refrigeration again desirable.

The consumer is by experience well acquainted with the usual signs of microbial spoilage in foods. Unfortunately, radiation pasteurized foods often lack these well-known warning signals.

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COMBINATION processes are here regarded as procedures whose influence is exerted primarily during the storage life of the food after irradiation: as distinct from factors which simply affect radiation inactivation as such (described in the previous papers in this section). Some processes, for example addition of ascorbic acid or cooking before irradiation, might obviously come into both categories—nevertheless the distinction helps to clarify our ideas.

The ideal objective of irradiation is to make possible the prolonged preservation of raw foods in their original fresh condition. Combination processes may thus be conveniently divided into two groups: first those compatible with that objective, such as storage under refrigeration, vacuum-packing, or addition of preservatives like antibiotics; and second, those which more or less alter the fresh nature of the food, like curing or heating. These five processes are all that there is time now to consider. Attention will of course be centred on the microbiological considerations, though there are others too; and discussion must be brief and simplified.

Used in the normal way, i.e. apart from irradiation, all the above processes have two fundamental features:

(a) They prevent or delay the development of the micro-organisms which usually cause spoilage and so determine the normal storage life.[1]

(b) They produce conditions more or less inimical to dangerous pathogenic organisms which, in the present context, are chiefly the following: *Cl. botulinum*, for which the "sterilizing" dose of radiation approaches 5 Mrads,[2] and the rather similar *Cl. welchii*;[3] enterotoxin-forming *Staphylococci*—the sterilizing dose probably about 0·75

Mrads;[4] and *Salmonellas*—sterilizing dose about 0·5 Mrads.[5]

The subject to be considered is, in essence, how far the virtues in these respects of a particular combination process may be useful to offset deficiencies in a simple process of irradiation, and vice versa.

*Refrigeration*

If food is to be stored frozen, there is no need to irradiate it simply to prolong its storage life: there is no danger from micro-organisms;[6] and the irradiation is likely indeed to accelerate development of rancidity in the fat.[7] Nevertheless, irradiation of a frozen food may be useful to destroy pathogenic bacteria (e.g. salmonellas in frozen egg)[8] or to "clean up" the material so that it has a longer life after thawing. Irradiation is the only process which can achieve these things with a frozen food.

To irradiate in the frozen state diminishes the effects of the irradiation; as explained by HORNE and BRIDGES in the previous paper; but this is directly related to the act of irradiation, and is not a combination process as here defined.

Refrigeration however also includes "chilled" storage, at 0 to 5°C, a procedure which by itself confers a storage life of a week or two for perishable foods like egg pulp, fish, meat or milk. It is well known that this life is longer the smaller the number of micro-organisms present initially, but that a quite substantial minimum is set to this number by what is hygienically practicable.[1] The virtue of irradiation is to permit a substantial further reduction in this initial number.

Reduction of the temperature below 5°C short of freezing has no significant influence on the process of irradiation as such; but, at those temperatures, spoilage of the above mentioned perishable foods is normally caused by bacteria which are fortunately very sensitive to irradiation,[9] so that their numbers can be reduced to low levels by moderate doses of radiation insufficient to cause appreciable changes in quality. Because the cold-tolerant micro-organisms are specially sensitive to irradiation, its benefits are generally much greater if the food is thereafter kept cool.

Pathogenic bacteria do not develop below 5°C;[6] though insufficient radiation may be used to kill them all, the low temperature protects against them. There is an obvious danger in proposals which have been made, to store incompletely sterilized foods at temperatures not low enough to effect this with certainty.

This then, is the basis of the "radiation–pasteurization" type of process; and the reason why it is recommended that such a process should be coupled with storage at chill temperatures.

In addition, of course, refrigeration delays changes in quality caused by enzyme and chemical reactions.

*Vacuum packing*

The mere removal of oxygen does not suffice to preserve perishable foods kept at normal temperatures, because the dangerous anaerobic bacteria can still grow freely in such circumstances. However, these bacteria do not grow below 5°C; while the usual micro-organisms which grow below 5° are greatly hindered when oxygen is absent.[10] The combination of low temperature and lack of oxygen is thus an effective one, and it has for example recently been proposed to ship meat in chilled holds filled with nitrogen.

Packing without oxygen is accordingly an obvious adjunct to radiation pasteurization with chilling, since the food must anyway be wrapped to prevent its re-contamination. How much oxygen might be allowed to remain is not yet known—some foods (e.g. fresh meat) absorb it,[11] and for them packing under vacuum as practised commercially might be equivalent to thorough gas-packing with nitrogen or carbon dioxide, and is much cheaper.

With meat treated with this combination of processes—i.e. irradiated, packed anaerobically and held at 5°C—a storage life of several weeks has been achieved with quite small doses of radiation (order of 0·25 Mrad).[12] The ultimate spoilage was however caused by unusual bacteria, among

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which faecal streptococci were prominent; so that this system, which has been little investigated, requires further study.

*Antibiotics and other preservatives*

These are substances effective in such small amounts that they produce no appreciable change in the food. Their action is to inhibit one or more of the groups of spoilage micro-organisms.

Antibiotics are especially interesting. The most promising are the tetracyclines which, used alone, increase the storage life of fish or meats, up to 100 per cent under favourable circumstances.[13] The choice of antibiotics is large, and their combination with irradiation has as yet been very scantily explored, except for the tetracyclines.

So far as is known, they do not modify the effects of radiation directly (as they sometimes do with heating e.g. nisin). Moreover, the tetracyclines do not kill the bacteria: they merely prevent particular types from multiplying. Thus, their action is simply to modify the nature of the micro-flora which develops after irradiation.

It is nevertheless established for chilled meats stored in air that irradiation coupled with treatment with tetracyclines gives an extension of storage life longer than with either alone: indeed, the respective extensions are roughly additive.[14,15] This seems to occur because some of the spoilage bacteria important in these circumstances, and able to survive the radiation, happen to be sensitive to the tetracyclines.[15]

Salmonellas resist the tetracyclines alone, but are not especially resistant to radiation. *Cl. botulinum* unfortunately resists both: hence it remains desirable to keep at chill temperatures below 5°C products like meat subjected to non-sterilizing irradiation + antibiotic treatments.

Yeasts and moulds are also relatively resistant to both treatments: hence they tend to dominate the flora of foods processed in this way, and an entirely abnormal kind of spoilage ensues.[14,15,16] This is prevented if, in addition, sorbic acid is added.[14]

Antibiotics may be regarded as a special case of the general class of antimicrobial additives to foods, of which there are many.[17] From the microbiological point of view one may ask for example, whether $SO_2$ added to fruit products or to (British) sausages will have any influence on their preservation by radiation. Such questions have yet to be investigated, and the field is wide. There are obvious medico–legal problems, too: besides doubts about irradiation itself, the addition of foreign substances to foods is regulated by laws, based on misgivings about the effect of such additives on public health, misgivings which apply not least to antibiotics; the combination of the two raises untold complications.

*Salting, etc.*

A cured product is, so to speak, one degree removed from the fresh state. Suitably cured products can ordinarily be kept for long periods at normal temperatures. Their composition makes them safe from salmonellas and botulism; while staphylococci are usually excluded, conceivably—though this is not certain—by the pressure of competing species.

Some cured products (notably U. S. bacon) withstand sterilizing doses of radiation without serious deterioration.[18] These are among the few foods for which, with present knowledge, irradiation could conceivably permit unrestricted handling under normal conditions. But apparently similar products (e.g. British bacon) may be severely damaged by such doses, for reasons unknown. With them, only pasteurization treatments are now possible, though these suffice to increase the storage life considerably. As refrigeration is not essential normally, it might be thought unnecessary for irradiated material likewise. It seems possible, however, that staphylococci surviving the irradiation (to which they may be relatively resistant) might develop at normal temperatures because the competing species were inhibited: this possibility needs investigation.

*Heating*

Heating may be applied before or after irradiation. Such treatment has three advantages:

(i) it greatly reduces the number of micro-organisms;

(ii) in appropriate circumstances, it has a sensitizing action for radiation (as HORNE and BRIDGES described);

(iii) it destroys enzymes, and so prevents subsequent changes (e.g. autolysis) which they might otherwise cause.

It has one obvious disadvantage—the food is no longer raw—for the above benefits are only achieved by the use of temperatures at which fresh foods become cooked.

It is common knowledge that it is unsafe to store cooked food for long, save in a refrigerator. This is because dangerous spore-forming organisms like *Cl. botulinum* and *Cl. welchii* survive cooking, unless undesirably prolonged. Most unfortunately, much the same organisms happen to be particularly resistant to radiation.[19] Therefore, cooking should be combined with either (a) irradiation sufficient to sterilize, or (b) failing this, pasteurization with refrigerated storage.

(a) The combination of heating with radiation is of interest in sterilization especially as offering the prospect of reducing the heat treatments needed to give a satisfactory degree of sterility to canned foods,[19,20] heat treatments which often cause considerable loss of quality. When heat-processing many canned foods however complete sterility is not aimed at, when it is known by experience that the properties of the food will keep the few surviving bacterial spores in a dormant condition ("commercial sterility"), though the precise reasons for this are often obscure.[21] If part of the inactivation in such a case is to be effected by irradiation, the process will be limited in application unless bacteria so treated remain equally dormant under equivalent conditions; there are suggestions that they may not, and the subject needs more study of a most difficult kind.

(b) Radiation-pasteurization of cooked foods, as noted, should entail refrigeration because normal cooking cannot be relied on to kill dangerous spores. The combined treatments would however produce near-sterile products, offering the prospect of refrigerator storage of ready-cooked foods over at least several weeks. This prospect has not yet been investigated extensively.

*New spoilage patterns*

As the object of all these processes is to eliminate the normal spoilage micro-flora, where there is not complete sterility the ultimate spoilage pattern is likely to be abnormal, as has already been shown in several cases.[14,15,16] The criteria which are common knowledge, for judging whether food is spoiled, then become valueless. Much new experience will have to be accumulated, not only by the technologist but also by the consumer, before a process with such an outcome is generally acceptable.

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# Summary of Recent Microbiological Studies in the U.S.A.

H. F. KRAYBILL*

Chemistry Division, U.S. Army Medical Research and Nutrition Laboratory, Denver 8, Colorado, U.S.A.

ONE objective of the American program is to use a non-pathogenic indicator organism for determination of the radio resistant properties of bacterial survivors in irradiated foods. Reference was made to the work of Anderson *et al.* (Oregon State College U.S.A.) on the radiation resistant micrococcus, a non-spore forming micro-organism which was found present in meat products treated at levels of 6 Mrep. This organism can be destroyed by mild heat or controlled by adjustment of salt content or chemical additives. Therefore the combination of mild heat treatment with irradiation has been suggested as a means of microbiological control for the radio resistant organisms.

Work at the Quartermaster Food and Container Institute for the Armed Forces (Chicago, Ill. U.S.A.) has revealed that with *Clostridium botulinum* spores (10,000 spores per tube) in an atmosphere of nitrogen none of the test organisms survived 1·5 Mrad. *Clostridium botulinum* spores were prevented in cheese at $1 \times 10^6$ rep; however, the product was not palatable and this radiation dose strain *T* type A of this organism in phosphates buffer required 286,600 rads for the *D* value (over 90% kill) and strain 62A required 223,000 rads. The *D* value for 115 B required about 223,000 rads. Four other strains studied required less radiation. For example 51 B had a *D* value of 129,300 rads.

Vegetative cells are more sensitive to radiation than spores. *D* values for spores in food products are higher because of protective action of the food material. For example the *D* values in buffer and in food for *Clostridium botulinum* spores are as follows:

D *Values on radiation for* Clostridium botulinum *spores*

| Mixed inocula in phosphate buffer | Mixed inocula in cream chicken soup |
|---|---|
| 232,500 rads | 352,000 rads |

The concept at present is that the high dosage of 4·5 Mrads may not be required for *Clostridium botulinum* as previously suggested for all foods since acid foods (below pH of 4·6) may require a much lower dosage.

* FAO Consultant on microbiology. Present address: Curtiss Wright Corporation, Princeton, New Jersey, U.S.A.

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# Discussion

Dr. Mossel (Netherlands): I should like to raise a point which may be of importance in evaluating the microbiological efficiency of the radiation treatment of foods. A group of workers has observed that a high proportion of bacterial cells in pure cultures cannot be cultured after irradiation, i.e. do not form colonies in the rich media and at the temperatures which are normally used for detecting or counting them; whereas they do at lower temperatures and in media less rich in known and unknown nutrients, and therefore not normally used for that purpose. Could not this phenomenon also occur in counting or subculturing survivors from irradiated foods? We could thus seriously overestimate the bactericidal efficiency of irradiation, for both pathogenic and food spoilage micro-organisms. If the meeting shares my misgivings, I think that we should, as a most urgent first step, carry out research on the various aspects of recovery after irradiation in order to revise present estimates of survival curves, if necessary.

Mme. Alibert (France): The influence of the culture medium on restoration after irradiation may not be a general phenomenon. Although Stapleton obtained with *E. coli* B/r a higher survival on enriched media, Miss Alper, on the contrary, obtained a higher survival on simple synthetic medium (with *E. coli* B, I believe). Personally I have not noticed any differences in survival after irradiation (with *serratia indica*) that were due to differences in the media, but the time elapsing between irradiation and the measurement can be important.

Mr. Bridges (U.K.): At the Wantage Radiation Laboratories we have confirmed the results of Stapleton (*E. coli* B/r) and Alper (*E. coli* B) concerning the effect of culture medium on survival of micro-organisms after irradiation. Several other Enterobacteriaceae have been found to behave in a similar way to *E. coli* B/r, that is, a higher survival is obtained using rich culture media. It is our opinion that *E. coli* B is the exception rather than the rule. However, we agree with Dr. Mossel that the possibility of pathogenic organisms surviving after irradiation should be investigated. American work with spore-forming organisms has shown that after irradiation in the spore state no post-irradiation medium effect is exhibited[1,2] but if they are irradiated in the vegetative state then an effect may be observed.[1] Since sterilizing treatments are mainly concerned with the elimination of spore-forming bacteria these investigations suggest that our present assessment of the sterilizing dose may be valid.

Dr. Roberts (U.K.): It must be borne in mind that the dose rate may have some influence on the killing of bacteria. Investigations with polymers of high molecular weight suggest that dose rate could play an important part in the inactivation of large biological molecules.

Dr. Laser (U.K.): I have done only a little work on the effect of dose rate on the inactivation of enzymes, but in the few cases in which dose rate has been varied within modest limits, no differences have been observed.

Professor Manil (Belgium): I should like to raise two questions. First, has the possibility of the development of radiation resistant micro-organisms been adequately considered? Secondly, I think it is desirable that the microbiologists present should meet to exchange information on their work.

Professor Pijanowski (Poland): I also would like to know more about the possibility of radiation resistant micro-organisms developing. With low dose treatment, particularly, this problem might arise. There are two aspects of this question:

(1) the slow development of an increased resistance to radiation
(2) the mutation induced by radiation might lead to the development of resistant organisms.

I should also like to ask Dr. Ingram if he knows whether bacterial toxins which might sometimes be present in material subjected to irradiation (for example meat and milk) are resistant to radiation.

Dr. URI (U.K.): I do not think that application of radiation sterilization on a large scale would lead to radiation resistant strains of micro-organisms in the absolute sense. However, one could contemplate a very gradual and slow increase in the lethal dose, due to survival of micro-organisms which have a larger concentration of protective substances in their cells. As far as the effect of dose rate is concerned, I have already emphasized its importance, and further information on the survival of micro-organisms as a function of dose rate, particularly at very high dose rates would be valuable. It would be interesting to have the views of microbiologists. Could Dr. Ingram comment on these points?

Dr. HANNAN (U.K.): With reference to the inactivation of *Clostridium botulinum* as quoted in the paper, I think it should be mentioned that a survival of one organism per $10^8$ g would not be acceptable for commercial practice since this represents the approximate daily output of only one cannery. A further safety factor of about a million would seem to be indicated for practical use.

Dr. HEEN (Norway): Is anything known about the effect of radiation on the antigenic mechanisms? Does an inactivation dose impair the antigen formation, or is a vaccine formed?

Dr. INGRAM (U.K.): Although there is little evidence that radiation resistant bacteria develop, the possibility of their doing so must most definitely be considered as with antibiotics. American work has shown that toxins are partially destroyed by radiation but their presence in food before irradiation would seem highly undesirable. I would refer Dr. Heen to Dr. Kraybill for work on antigen formation.

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# 3. PRESENT STATUS OF RADIATION PRESERVATION OF FOODS

(a) Meat, poultry and fish

(b) Eggs and egg products

(c) Milk and milk products

(d) Potatoes and root crops

(e) Fresh fruits and vegetables

(f) Disinfestation of food

(g) Packaging

(h) Economic considerations of food irradiation

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# The Effects of Irradiation on the Quality of Meat and Poultry

B. COLEBY

Low Temperature Station for Research in Biochemistry and Biophysics,
Department of Scientific and Industrial Research, Cambridge

Although different meats vary considerably in their response to irradiation, the changes in colour, odour and flavour produced with sterilizing doses (4·8 Mrads) are usually sufficiently pronounced to render the process of doubtful commercial value. Pasteurizing doses of radiation (up to 1 Mrad) appreciably extend the time required for microbial spoilage of meats under chilled storage, and in many cases, such doses produce little or no off-flavours.

With both high and low dose treatments, the chemical and physical environment of the meat has an important influence on these adverse changes, and the effects of removing oxygen and freezing are discussed.

Storage of irradiated meats usually results in a further loss of quality. With sterilized meats this proceeds more rapidly at high temperatures; and with pasteurized meats, flavour deterioration is often noticeable before microbial spoilage occurs. In many cases, this deterioration is due to oxidative rancidity of lipids, but this is not always the explanation. Pasteurization processes appear to give a small net increase in the period for which meats of high quality can be stored under chilled conditions.

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ANY method which potentially gives an extension of the storage life of meat and poultry has attractions, and is worthy of investigation. Meat products are comparatively expensive and, for the most part, have a very limited shelf life. Even under chilled conditions of storage, a cut of meat cannot usually be kept for longer than 2 weeks.

Conventional methods of preservation all have their characteristic limitations. Heat processing—obviously—cannot preserve meat in its raw state, and while the products are usually of attractive flavour, they differ considerably from the starting material. Also, the desired degree of bacterial inactivation often results in some overcooking of the meat. Freezing, on the other hand, permits of the extended storage of meat, with excellent retention of its original characteristics and nutritional properties. Some change of colour occurs, however, and subsequent thawing leads to exudation of fluid ("drip"), causing slight consumer resistance. This detracts from the process.

Potentially, radiation preservation has certain advantages. There is the prospect of preserving raw meat for prolonged periods; the process is eminently suitable for continuous processing; the radiations are sufficiently penetrating to treat large pieces of meat; and it might be possible to use plastic film packaging materials as prolonged processing at high temperatures is avoided. It is easy to visualize applications at all stages in the handling of the meat, from the slaughter house to the domestic refrigerator, though there is general agreement that radiation processing would be more useful in highly organized societies with well recognized food distribution channels.

As with many fields of food science, there have been conflicting reports about the benefits to be achieved through irradiation. Partly this is due to the well known difficulties in obtaining a uniform starting material, but much of it is because of the important role that physical conditions, both during and after irradiation, can play in determining the quality of irradiated meat products. However, the broad picture is becoming clearer now.

## THE EFFECTS OF STERILIZING DOSES OF RADIATION

The most attractive potentiality for using irradiation is the complete elimination of micro-organisms. Although sterile, such meat products would not have indefinite life at elevated temperatures since some enzymic and chemical changes continue; nevertheless, the absence of microbial spoilage would be a tremendous advantage. As has been pointed out by HORNE and BRIDGES,[1] the radiation dose necessary for "commercial" sterility may vary according to the criteria one uses to evaluate it, but for meat products of high pH, a dose of about 4·8 Mrads is indicated. Unfortunately, at this dose level, the chemical changes associated with irradiation tend to have adverse effects on many meats. The colour can be markedly affected, and changes in odour and flavour are usually noticeable. Descriptions of the odour and flavour changes vary, but most people regard them as unattractive. Each meat responds differently; pork, chicken, turkey and bacon are less affected than lamb and beef. The reasons for this are not understood at present. But even with those products which are least affected, it seems doubtful if the average consumer would find the product attractive. Although radiation sterilized meats may find some specific application, e.g. for military purposes, it is difficult to foresee the general acceptance on the civilian market of meats sterilized by radiation with present techniques. It has been suggested that consumers might not reject radiation sterilized meats merely because of the unusual nature of the flavours due to irradiation;[2] but until irradiation is a well established process it would seem safer to assume that consumers buy food on the assumption that it will taste as it has customarily tasted.

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## THE USE OF RADIATION PASTEURIZATION

Because of the adverse changes associated with sterilizing doses of radiation, there is an increasing interest in the advantages that might be obtained using lower doses of radiation. The combination of heat and irradiation can give sterile products, but as INGRAM[3] has stressed, there is need for much experience before the microbiological aspects of such processes can be evaluated. Rather more experience has been gained in the use of radiation pasteurization techniques, i.e. the storage at chilled temperatures of meats which have received moderate doses of radiation. There is general agreement[2,4] that meat stored at 0° to 5°C after being irradiated with doses in the range 50,000 to 1,000,000 rads can be held between five and ten times as long as unirradiated meat before microbial spoilage terminates the shelf life. It is usually possible to choose a dose which results in only very slight flavour changes, while appreciably extending the storage life. There seems to be a particular dose below which little useful increase is achieved: in the case of minced chicken meat stored anaerobically, this dose is about 250,000 rads.[5] At about this level of radiation, off-flavours just become detectable if the chicken meat is cooked by steaming.

The threshold dose at which quality changes can be distinguished depends upon the method of assessment. The odour of the raw meat is one of the most sensitive sensory tests, and it is often possible to detect changes in odour after doses as low as 50,000 rads. Cooking usually makes it difficult or impossible to detect such low doses. The experiments of HANNAN and SHEPHERD[6] illustrate this point: after even light cooking, a dose in excess of 250,000 rads was necessary before discrimination could be made using minced chicken meat. Even higher doses of radiation can be used with harsher cooking methods, as is illustrated by the data in Table 1, which show discrimination between irradiated chicken meat samples when the meat is cooked by steaming, but lack of discrimination among similar samples cooked by roasting. In fact, with whole chickens, cooked by the normal roasting method, a dose of 800,000 rads produces scarcely detectable flavour changes; and such a dose will

TABLE 1. Taste panel assessment of whole eviscerated chickens irradiated with cobalt 60 $\gamma$-rays

| Sample | Method of cooking | Average rank for doses (rads) of: | | | | No. on panel |
|---|---|---|---|---|---|---|
| | | 0 | 125,000 | 250,000 | 575,000 | |
| Whole carcasses | Odour of raw bird | 1·0 | 2·4 | 3·2 | 3·4 | 5 |
| Breast meat | Cooked in portions by steaming | 1·8 | 2·1 | 2·8 | 3·3 | 10 |
| Thigh meat | | 1·8 | 2·6 | 3·8 | 2·8 | 5 |
| Drumstick | | 1·6 | 1·8 | 3·2 | 3·4 | 5 |
| Breast meat | Bird roasted whole | 2·7 | 2·3 | 2·5 | 2·5 | 10 |
| Thigh meat | | 2·2 | 2·0 | 2·6 | 3·2 | 6 |
| Drumstick | | 2·4 | 2·8 | 3·0 | 1·8 | 6 |

The judges were asked to rank the samples in order of preference. A low average rank indicates a preferred sample and a high average rank a disliked sample; samples which cannot be differentiated have similar average ranks.

increase the storage life of a chicken held at 1° from about 10–12 days to 60 days before microbial spoilage occurs. The relative threshold levels for the detection of irradiation induced flavours vary for different meats; and criteria can obviously be strongly influenced by the eventual destination of the product, since meat intended for use in restaurants or hotels will be cooked before reaching the consumer, who will thus be spared any unpleasant odours evolved during cooking.

## THE EFFECTS OF CHEMICAL AND PHYSICAL ENVIRONMENT

At both high and low levels of radiation the chemical and physical environment of the meat can profoundly modify the quality changes during both the irradiation and the subsequent storage.[7] The presence or absence of oxygen is a variable which has attracted considerable attention. While oxygen is desirable to achieve a high microbiological kill,[1] it has commonly been reported that quality changes are more adverse in its presence.[8] Certainly, colour changes are more adverse: irradiation in the presence of oxygen tends to give brown products due to the formation of metmyoglobin, whereas irradiation in the absence of oxygen usually confers an attractive red coloration. Contrary to common experience, experiments at the Low Temperature Research Station have shown that odours and flavours of meats irradiated in the presence of oxygen are often preferred to those irradiated *in vacuo*. This is particularly so in the case of chicken; in the case of minced beef, more variable results have been obtained (Table 2). While differences can be detected fairly readily it is less easy to decide which is the preferred treatment. Sometimes the choice is reversed by the act of cooking. Probably the differences are rather small. In addition, it is difficult to determine how the oxidation–reduction potential in a piece of meat is affected by its exposure to oxygen.

The exclusion of oxygen during prolonged storage is clearly necessary to prevent changes due to oxidative rancidity in the fatty tissue of meat. Irradiation usually hastens the onset of oxidative deterioration and as LEA and MACFARLANE[9] have shown in the case of beef fat, doses as low as 100,000 rads can produce easily detectable irradiation flavours of an oxidative nature.

Quality changes also depend markedly upon whether the meat is frozen during irradiation. Odour, flavour and colour changes are normally much less with frozen

TABLE 2. The effect of the presence or absence of oxygen during irradiation on the ranking for preference by odour and flavour of chicken and beef samples

| Sample, test and conditions of irradiation | Dose (Mrad) | Number of times ranked | | | | Average | |
|---|---|---|---|---|---|---|---|
| | | 1st | 2nd | 3rd | 4th | Rank | Hedonic score* |
| *Chicken Breast* | | | | | | | |
| *Odour*† | | | | | | | |
| In air | 1 | 34 | 8 | 5 | 1 | 1·4 | — |
| In nitrogen | 1 | 10 | 22 | 13 | 3 | 2·2 | — |
| In air | 2 | 2 | 17 | 19 | 10 | 2·8 | — |
| In nitrogen | 2 | 2 | 1 | 11 | 34 | 3·6 | — |
| *Flavour*‡ | | | | | | | |
| In air | 1 | 3 | 4 | 0 | 1 | 1·9 | — |
| In nitrogen | 1 | 3 | 2 | 0 | 3 | 2·4 | — |
| In air | 2 | 0 | 1 | 5 | 2 | 3·1 | — |
| In nitrogen | 2 | 1 | 1 | 3 | 3 | 3·0 | — |
| *Minced Beef* | | | | | | | |
| *Odour*† | | | | | | | |
| In air | 1 | 1 | 13 | — | — | 2·0 | — |
| In nitrogen | 1 | 13 | 1 | — | — | 1·0 | — |
| *Flavour*‡ | | | | | | | |
| In air | 1 | 4 | 1 | — | — | 1·2 | 4·2 |
| In nitrogen | 1 | 1 | 4 | — | — | 1·8 | 4·2 |

* A nine point scale, with the mid point, 5, being rated "neither like nor dislike."
† Selection significant at 1% level.
‡ Selection not significant.

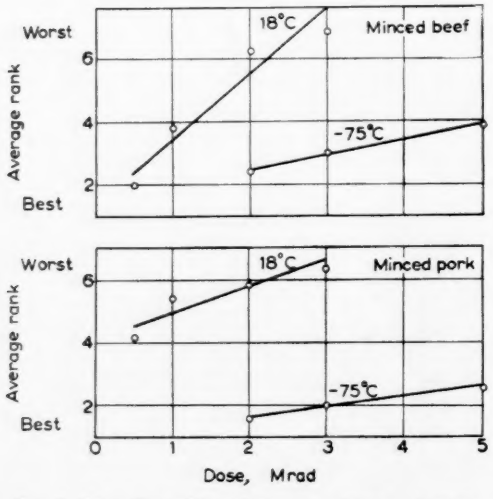


FIG. 1. The effect of irradiation at −75°C and 18°C on the ranking for odour of raw beef and pork samples by a five member panel.

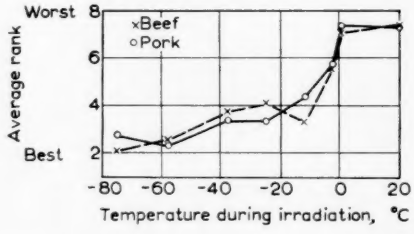


FIG. 2(a). Ranking for odour by 3–4 member panel of samples of minced beef and minced pork irradiated at various temperatures. Dose of 2·5 Mrads.

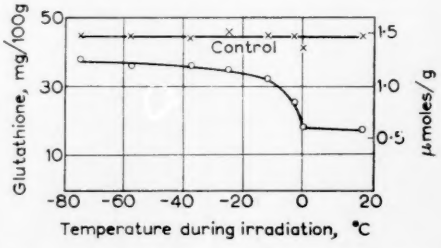


FIG. 2(b). The destruction of glutathione in minced pork irradiated with 2·5 Mrads at various temperatures.

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samples.[8] The data in Fig. 1 illustrate this effect with the odour of samples of beef and pork. It is quite clear that samples irradiated at −75°C are distinguished from those irradiated at room temperature. Furthermore, comparison of samples given different doses at these temperatures shows that beef samples irradiated at −75° can withstand a dose about four times as great as that which produces a similar odour in the raw meat irradiated at room temperature. With pork, the corresponding gain is by a factor greater than 10, samples given 5 Mrads at −75°C being preferred for odour to those given 500,000 rads at 18°C. Cooking tends to reduce the gain conferred by freezing, factors of about 1·5 and 4 being observed respectively for the flavour of beef and pork.

Further study of the temperature region in which the protection by freezing becomes effective reveals a rather abrupt transition in the range −3° to −6°C; similar results are obtained if a chemical criterion of radiation damage is used; e.g. the destruction of glutathione.[10] This is illustrated in Fig. 2. The exact temperature of this transition depends upon whether the meat is brought to the required temperature from above or below, which suggests that the effect depends upon the physical state of the water in the tissue.

## QUALITY CHANGES DURING STORAGE

The object of any method of food preservation is to prevent undesired changes in quality during storage. Irradiation must be considered singularly unsuccessful in this respect. Some reports suggest that prolonged storage of meats sterilized by radiation results in an improvement in quality.[11] In our experience this is not always so. The data in Table 3 show that minced beef sterilized by radiation and stored at 0°, 12° and 25°C undergoes a gradual deterioration on storage at all three temperatures. Furthermore, after a given period, the preferred sample was always that stored at the lowest temperature. In any case, the flavours, even initially, were very poor.

TABLE 3. The effect of storage at difference temperatures on the flavour of minced lean beef packed under nitrogen in aluminium cans and irradiated with 5 Mrads at −75°C

| Temperature of storage | Average hedonic rating* Days of storage | | |
|---|---|---|---|
| | 21 | 53 | 92 |
| 0°C | 4·6 | 4·1 | 3·5 |
| 12°C | 2·6 | 1·8 | 2·2 |
| 25°C | 2·0 | 1·8 | 1·9 |

* Assessment by five member panel using a nine point hedonic scale: 5 = neither like nor dislike; 2 = dislike very much.

It is not always appreciated that loss of quality, which is not due to microbial activity, can occur during the storage of radiation pasteurized meats. Thus, although chickens irradiated with 800,000 rads are preserved from spoilage by bacteria for more than 8 weeks at 1°C, the useful storage life before loss of quality becomes apparent is somewhat less than 3 weeks, i.e. about a 50 per cent gain on the storage life of unirradiated chickens. This is illustrated in Fig. 3. It is interesting to note that whereas

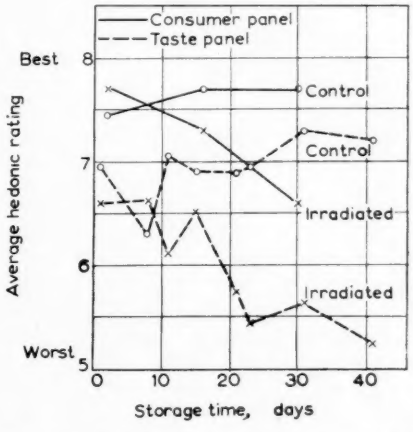


FIG. 3. Effect of storage at 1°C on flavour of chickens irradiated with 800,000 rads. Assessment by taste panel of 20 and consumer panel of 130.

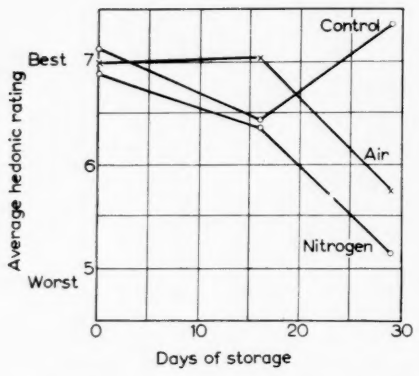


FIG. 4. The effects of storage at 1°C in air and nitrogen on the flavour of chickens irradiated with 700,000 rads. Assessment by a panel of 10.

the taste panel of ten always had a frozen control sample (undeclared) for comparison, the members of the consumer panel of 130 were given either a control chicken or an irradiated one, but not both together. In spite of this, the assessment by the taste panel corresponds strikingly with that of the consumer panel. It should be noted that the chickens do not become positively distasteful after storage for more than 3 weeks: they merely become rather insipid and are of obviously inferior quality. One report[12] suggests that rancid flavours contribute to this deterioration; our experience, on the contrary, shows that chickens stored with access to air are preferred to those stored in nitrogen even 30 days after irradiation (Fig. 4).

Sausages irradiated with 1 Mrad also show flavour deterioration on storage at 1°C, although in this case the samples stored in the absence of oxygen were preferred for a while (Fig. 5). Eventually, flavours of a rancid kind developed, due to oxygen penetrating the plastic film pack.

Such flavour deterioration during storage is an aspect of radiation pasteurization which is not often emphasized, and which detracts from the apparent benefits to be gained.

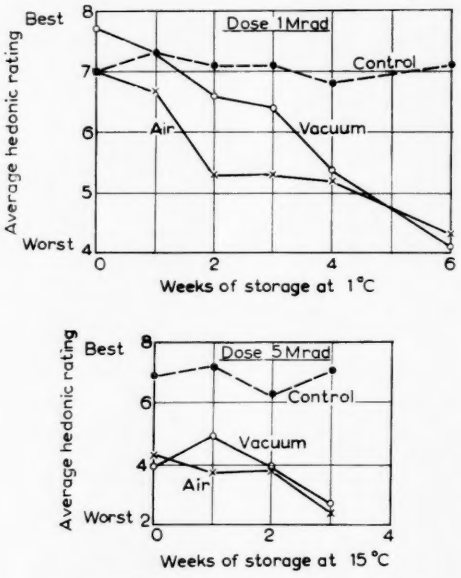


FIG. 5. The effects of storage on the flavour of irradiated sausages packed in cellophane/polyethylene film. Assessment by a panel of 10.

## PRESENT OUTLOOK

In summary, one may say that the prospects for general application of the radiation sterilization of meats appear remote at present; and that even the useful increase in storage life obtained using radiation pasteurization techniques may be more limited than is sometimes supposed. There would appear, though, to be some real gain in using pasteurization processes, and this may be of value in the distribution of meats. It should be noted, however, that our experiments have never given us an irradiated meat which was as good as a corresponding sample which had been stored in the frozen state. The benefits of irradiation are such that they will be appreciated by the food specialist long before they are by the man in the street, who tends to lose interest when he learns that irradiated meats cannot be stored on the shelf for ever. Much more work needs to be done before the possible applications can be evaluated: the effects of oxygen, other than in producing rancidity, cannot be explained or predicted; that of

freezing during irradiation is ill-defined; and there is disagreement about the benefits of storage. Technological aspects need particular attention: e.g. irradiation often causes the exudation of fluid ("weep"), and if this exceeds 1 or 2 per cent of the weight of the meat, then the economic implications may be serious, to judge from experience with frozen meat. Since the benefits of radiation processing of meats appear rather limited, economic factors will be of prime importance in determining its future exploitation.

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# Changes in Spoilage Pattern of Chicken Meat as a Result of Irradiation

M. INGRAM and MARGARET J. THORNLEY

Low Temperature Station for Research in Biochemistry and Biophysics,
Department of Scientific and Industrial Research, Cambridge

The microflora of radiation-pasteurized chicken meat is described in detail: it broadly resembles that reported by American workers for beef.

With anaerobic minced meat kept at 5°C, doses of 0·25 Mrad delayed spoilage for several weeks. The ultimate spoilage flora consisted of microbacteria and faecal streptococci; the enrichment of the latter raises misgivings about this type of process.

With whole eviscerated chickens, stored at 1°–3°C in loose film bags, doses up to 0·8 Mrad increased the storage life roughly from 10 to 50 days. The spoilage microflora was somewhat different in different experiments, and corresponding differences were observed in the off-odours which heralded spoilage. In all cases, however, the principal organisms were bacteria of types common on chilled meat.

It is concluded that there are no special bacteriological indications against radiation pasteurized meats, provided they are packed in air and kept at chill temperatures.

ANY particular food is normally spoiled by a characteristic association of micro-organisms, whose activities ultimately produce typical changes in appearance, odour or taste; and experience has taught the consumer to recognize these changes as signs that the food is no longer fit to use. Irradiation alters these characteristic patterns, and so removes the normal warning signs. Hence it is important to investigate the nature of the alterations.

Such investigations are laborious, and ours have so far dealt with only one set of circumstances, which merit brief explanation.

(a) We have worked only with chicken meat, because this suffers relatively little organoleptic damage on irradiation.

(b) We have used doses insufficient to sterilize, because sterilizing doses—even with chicken—produce such damage to an undesirable degree.

(c) We have stored at temperatures below 5°C because, at higher temperatures, the possibility that pathogenic organisms might develop would necessitate a degree of control which we cannot yet exercise.

Within these limits, we have studied two broadly different systems.

(i) The chicken meat from all of the carcasses, without bones or skin, was minced and packed under nitrogen in shallow aluminium cans, depth 1 cm capacity 30 g. In this system, micro-organisms are distributed throughout the mass; and conditions probably become anaerobic rapidly, through the consumption of residual oxygen by the tissues—the samples were stored at 0°C overnight before irradiation, to allow this to take place. The samples were irradiated with 4 MeV cathode rays. The organoleptic results of this treatment have already been described.

(ii) Eviscerated whole carcasses were used. Here spoilage is caused by the development on the surfaces of a slime of aerobic or facultatively anaerobic bacteria, chiefly derived—as with other meats—from surface contamination during handling. The carcasses were wrapped in sealed but loose polythene bags, to prevent misleading contamination subsequently; this created conditions of high humidity, and may have partly restricted access of oxygen. Cobalt-60 gamma rays were used for irradiation.

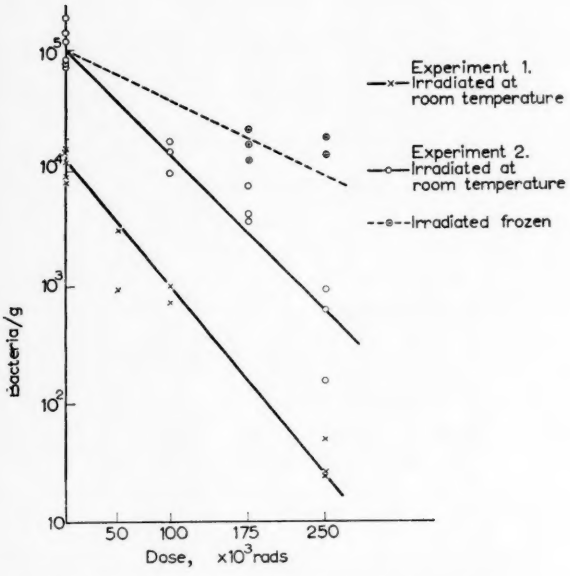


FIG. 1. The effect of irradiation with 4 MeV cathode rays on the survival of bacteria in minced chicken meat.

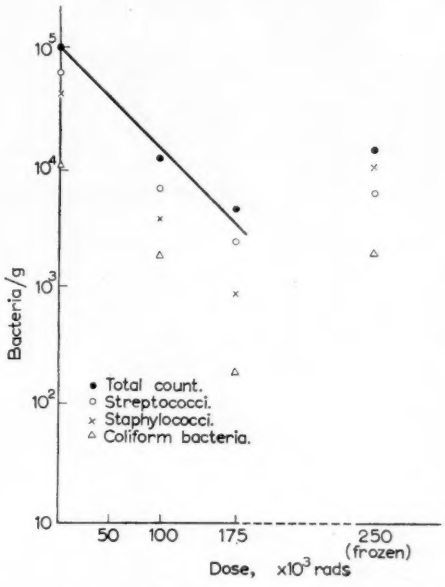


FIG. 2. The effect of irradiation with 4 MeV cathode rays on the survival of different types of bacteria in minced chicken meat.

The canned minced chicken will be considered first.

Control material contained about $10^5$ bacteria/g—a comparatively small number for minced meat, since this was prepared in a hygienic manner, in the laboratory. After 14 days at 5°C, numbers had increased to about $10^8$/g and the meat stank, behaviour similar to that of other meats. The nature of the bacteria will be described below.

The numbers of bacteria at the outset of storage were reduced by irradiation, the diminution being roughly exponential with dose over the range used, which is the classical picture (Fig. 1). There was, moreover, no clear evidence that the radiation exerted any selective action between the groups of bacteria which were distinguished (Fig. 2). If the samples were frozen at about −60°C for irradiation, the dose needed to achieve a given kill was increased $2\frac{1}{2}$ times—this factor is roughly the same as for organoleptic changes.

The storage life at 5°C increased progressively with the dose of radiation, though with 100 krads the increase was trifling—about 3 days (Fig. 3). With 175 krads, different cans spoiled at very different times, some still being sound after 3 weeks. With

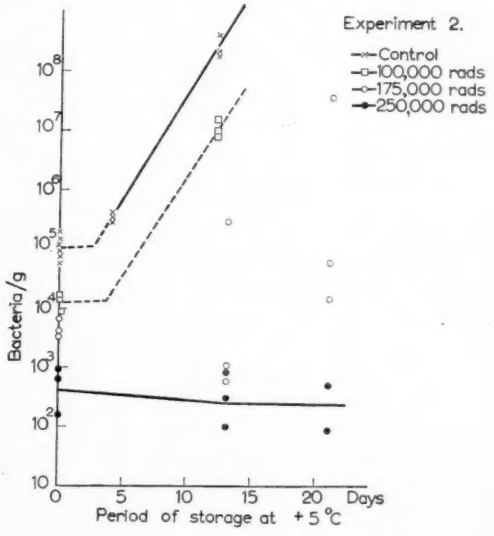


FIG. 3 The effect of storage at 5°C on the viable count of control and irradiated samples of minced chicken meat.

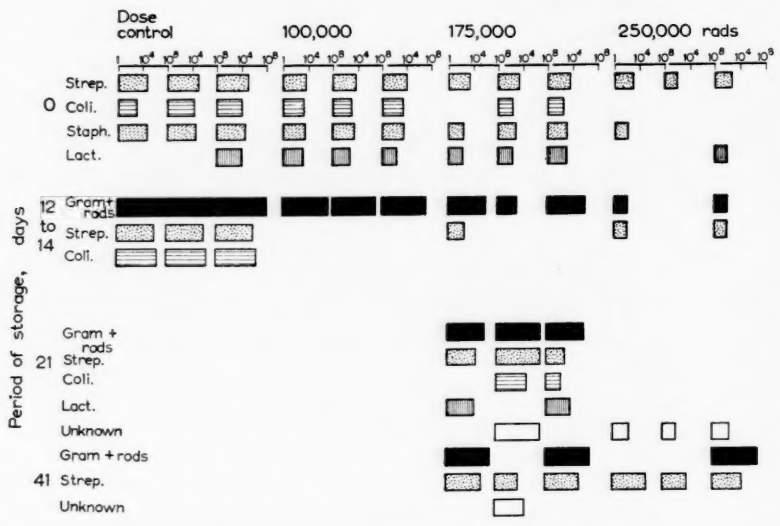


FIG. 4. The effect of storage at 5°C on the numbers of different types of bacteria in control and irradiated samples of minced chicken meat.

250 krads, little change occurred during 3 weeks, but the bacteria had begun to develop in some cans after 6 weeks, and after 43 weeks only one can out of 6 showed little change.

This is shown in Fig. 4, which reviews the analysis of the nature of the bacteria. In the control meat, the striking feature is the replacement of the initial flora by a spoilage flora consisting predominantly of lactobacilli and Gram-negative rods, with some faecal streptococci, a reflection of the near anaerobic conditions prevailing: the absence of strict anaerobes may be ascribed to the low temperatures. With the highest dosage, 250 krads, the Gram-negative rods had apparently been completely eliminated; and the lactobacilli almost so, for they were absent from 2 of the 3 cans examined towards the end of their storage life, though dominant in the one can where they were present. These lactic acid bacteria appearing on storage in the irradiated samples were somewhat different from those in the controls: they belong to the genus *Microbacterium* and resemble organisms isolated at the American Meat Institute from irradiated beef.[1]

The odours of the spoiling irradiated samples were not noticeably different from those of controls—described as putrid or faecal (besides the irradiation odour); which is, perhaps, not surprising as rather similar lactobacilli predominated in most cases. Two points may be mentioned: the same odour was several times detected in cans with comparatively low bacterial counts (order of $10^4$–$10^5$/g); and nothing unusual was noted in the two cans (250 krads, 21 days) where streptococci predominated. No greening was observed in any of these samples.

Let us turn now to the whole chickens. The increases in storage life following different doses of irradiation are shown in Fig. 5: with the higher doses the increase is substantial, greater than has been attained with chlortetracycline alone. The effects of CTC and irradiation were approximately supplementary. The length of the storage life was determined by briefly opening the bags and smelling the birds, taking care not to introduce contamination.

The flora on control birds consisted of a mixture of coliform bacteria, *Achromobacter* and non-pigmented *Pseudomonas* strains. During storage at +3°C, the latter group (which used to be, and still are called Achromobacter by many French workers e.g. BRISOU[2])

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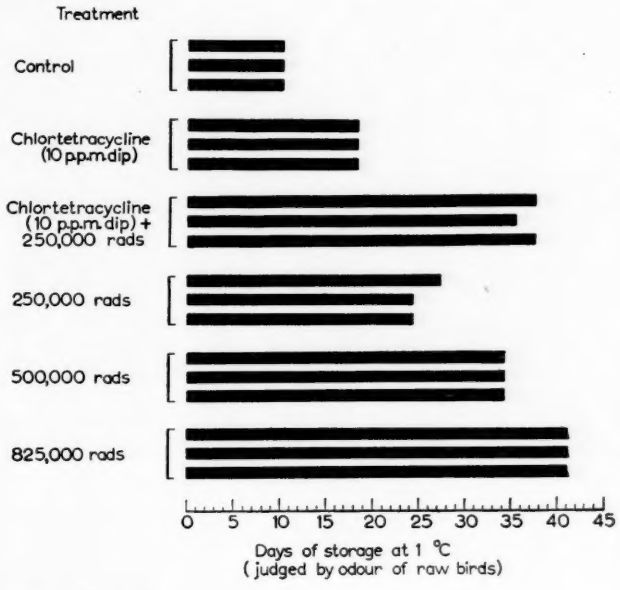
Treatment
Control
Chlortetracycline (10 p.p.m. dip)
Chlortetracycline (10 p.p.m. dip) + 250,000 rads
250,000 rads
500,000 rads
825,000 rads
0 5 10 15 20 25 30 35 40 45
Days of storage at 1 °C
( judged by odour of raw birds)

FIG. 5. The effects of chlortetracycline and irradiation with cobalt-60 gamma rays on the storage life of whole eviscerated chicken at 1°C.

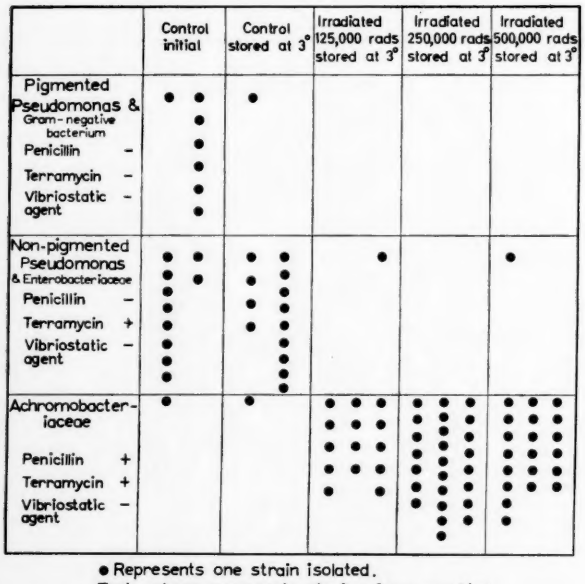


FIG. 6. The effects of irradiation with cobalt-60 gamma rays and storage at 3°C on the types of bacteria present on eviscerated chicken carcasses, as indicated by the antibiotic sensitivity test of SHEWAN *et al.*[3]

became almost wholly predominant (Fig. 6) with the development of a typical putrid smell. The situation is broadly the same as with other meats.

Irradiated samples behaved differently. In our first experiment they developed a quite different odour—sweetish, not so strong, and developing gradually so that it was more difficult to decide precisely when a sample should be adjudged spoiled. The spoilage flora consisted almost wholly of *Achromobacter* (Fig. 6). This agrees with American suggestions that *Pseudomonas* are unusually readily killed by irradiation.[1]

It also illuminates two other features of the situation. First, a reason why CTC supplements irradiation is probably that the important organisms resisting irradiation are sensitive to tetracyclines. Second, subsequent work has suggested that strains of *Achromobacter* are less active than *Pseudomonas* in producing amine-like compounds, especially with restricted access of air; which may be why, as agents of spoilage, they produce less putrid smells.

A similar experiment, with a rather higher dose, gave an essentially similar result—a preponderance of *Achromobacter* on the irradiated birds (Table 1) and sweetish odours on spoilage. There were some differences: the flora on the control birds was more diverse including a significant Gram-positive element. Correspondingly, lactobacilli occurred on some of the irradiated birds: from the predominance of similar organisms under anaerobic conditions, this might be related to partial restriction of air supply by the wrappers.

A further experiment, still incomplete, is exploring the effects of somewhat higher doses, in more detail. At doses exceeding 250 krads, the survival curve ceased to be exponential with dose, indicating the survival of a somewhat more resistant group of organisms (Fig. 7). Detailed examination revealed that, as before, the *Pseudomonas* were the first to be eliminated by small doses below 250 krads, and that the population surviving 500 krads consisted mostly of yeasts (Table 2). The predominance of yeasts was, however, only temporary, for they had evidently been overwhelmed by bacteria in the final spoilage flora. (This does not happen in the presence of CTC, which the yeasts resist better than the bacteria.) A striking feature of this experiment was the predominance of non-pigmented *Pseudomonas* in the spoilage flora of the irradiated samples, though *Achromobacter* strains were common around the 250 krads level; while at 852 krads pigmented *Pseudomonas* appeared in quantity. Though the reasons for such differences from the earlier experiments are unknown (these carcasses

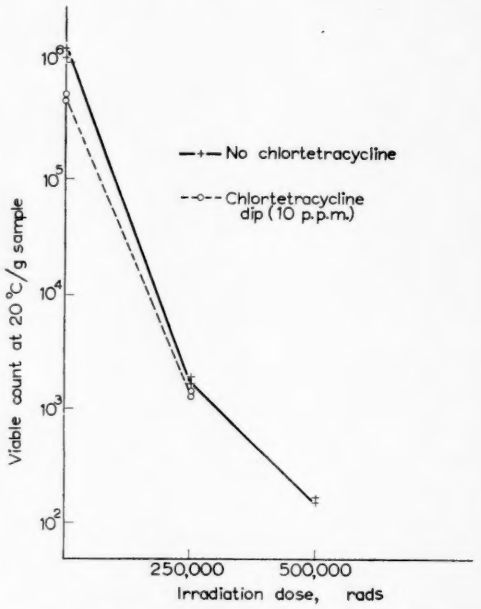


FIG. 7. The effects of chlortetracycline and irradiation with cobalt-60 gamma rays on the survival of bacteria in whole eviscerated chicken.

TABLE 1. The effect of irradiation with cobalt-60 gamma rays, and storage at 3°C, on the percentage composition of the microflora of chicken carcasses

| | Percentage colonies on 20°C plates for: | | |
|---|---|---|---|
| | Control initial | Control spoiled at 3°C | Irradiated with 575,000 rads spoiled at 3°C |
| Pigmented *Pseudomonas* | 3 | 13 | 4 |
| Non-pigmented *Pseudomonas* | 3 | 15 | — |
| Achromobacter | 8 | 50 | 77 |
| Gram-negative Gas-producers | 53 | 10 | — |
| Other Gram-negative bacteria | — | 12 | 3 |
| Lactobacilli | — | — | 12 |
| Other Gram-positive bacteria | 33 | — | 4 |
| Number of strains studied | 36 | 68 | 98 |

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TABLE 2. The effect of irradiation with cobalt-60 gamma rays, and storage at 1°C, on the percentage composition of the psychrophilic microflora of chicken carcasses

| | Percentage colonies on 1°C plates for: | | | | | | |
|---|---|---|---|---|---|---|---|
| | Initial samples | | | Spoiled samples | | | |
| | Control | Irradiated | | Control | Irradiated | | |
| | | 250,000 rads | 500,000 rads | | 250,000 rads | 500,000 rads | 825,000 rads |
| Pigmented *Pseudomonas* | 27 | — | — | 7 | 11 | 3 | 29 |
| Non-pigmented *Pseudomonas* | — | — | — | 64 | 52 | 77 | 71 |
| *Achromobacter* | 41 | 11 | — | 18 | 37 | 20 | — |
| Gram-negative Gas-producers | 9 | — | — | 11 | — | — | — |
| Other Gram-negative bacteria | 9 | — | — | — | — | — | — |
| Gram-positive bacteria | 14 | — | — | — | — | — | — |
| Yeasts* | — | 89 | 100 | — | — | — | — |
| Number of strains studied | 22 | 19 | 23 | 28 | 27 | 31 | 24 |

* Some picked also from 20°C plates.

came from a different slaughterhouse), the predominance of *Pseudomonas* was signalled—with control and irradiated birds alike—by putrid smells resembling those in the earlier experiments.

One naturally hopes to interpret the behaviour of different organisms during spoilage in terms of their physiological peculiarities. For example, the behaviour of the yeasts in the last experiment is readily understood because they are more resistant to radiation than bacteria, but have much lower rates of cell division. Again, the near elimination of non-pigmented *Pseudomonas* by modest doses of irradiation, with their rapid assumption of dominance during cool storage, are consistent with the known properties of such organisms and their presumed susceptibility to radiation. We naturally wish to know whether our *Achromobacter* strains are in general more resistant, whether the pigmented *Pseudomonas* prominent on the 825 krads samples are exceptions to the rule, whether the microbacteria and streptococci of the minced chicken are unusually resistant, and so on. Our experiments for this purpose are in progress now; and the only indication so far is that the *Achromobacter* strains of the irradiated birds do indeed possess a greater resistance than the other organisms involved.

The most important practical aspect of such investigations is their bearing on the safety or otherwise of meats treated in this manner. As regards the eviscerated whole carcasses, nothing untoward has yet appeared. The spoilage flora of the irradiated birds has consisted of non-pathogenic organisms similar to those which occur commonly on normal meats, under refrigeration, and the chief peculiarity is an unfamiliar smell towards the end of the storage life. With the canned minced meat, on the other hand, the prevalence of faecal streptococci—not signalled by any obviously unusual feature—raises some doubts; for where organisms of this group have grown on meat it has sometimes been suspected that food poisoning has ensued. This situation requires further study, but a desirable preliminary is a greater degree of precision in identifying the food poisoning streptococci.

It should be obvious that the above considerations would not apply to partially sterilized meats stored at higher temperatures. Without refrigeration there is the possibility that dangerous salmonellas, staphylococci, and clostridia might grow. Hence extensive

investigations, of a kind similar to those just described, will be necessary before radio-pasteurization without refrigeration can be recommended for meats.

*Acknowledgement*—This work formed part of the programme of the Low Temperature Research Station of the Department of Scientific and Industrial Research.

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# Irradiation de la Viande en Relation avec d'Autres Procédés de Conservation, plus Particulièrement l'Inhibition de l'Autolyse Enzymatique par l'Adrénaline*

C. RADOUCO-THOMAS†

La détérioration de la viande est intimement liée aux phénomènes d'autolyse et à la contamination par les micro-organismes. Ce travail décrit un nouveau procédé permettant une meilleure conservation de la viande en combinant l'effet anti-autolytique de l'adrénaline avec l'action anti-microbienne des irradiations.

Des échantillons de tissus musculaires d'animaux de laboratoire et d'abattoirs (rats, lapins, poulet, porc, agneau, boeuf) ont été contrôlés immédiatement post-mortem et pendant des conservations de plusieurs semaines, à différentes températures (4°C, 25°C, 38°C).

Pour les muscles adrénalisés, les modifications biochimiques, structurales et organoleptiques sont caractérisées par une forte inhibition de l'exsudation ("drip") et des phénomènes structuraux de désintégration. De plus, la chute caractéristique du pH post-mortem du muscle est inhibée, tandis que les caractères organoleptiques de la viande (particulièrement la tendreté) sont stabilisés ou améliorés.

Le traitement combiné (adrénaline/irradiation) a donné de bons résultats sur le porc, le boeuf et le poulet et ne semble pas rendre la viande impropre à la consommation.

Un tel procédé combiné, utilisé conjointement avec un emballage protecteur adéquat pourrait permettre une meilleure conservation de la viande aux températures de congélation, normales et tropicales. Le choix entre l'irradiation de surface at une irradiation pénétrante reste lié au problème de la stérilité du muscle.

Spoilage of meat is closely associated with autolysis and microbial development and the paper describes a new process for the better preservation of meat by combining an epinephrine anti-autolytic treatment with irradiation as an anti-microbial treatment. The anti-autolytic effect is produced by ante-mortem administration of epinephrine.

Samples of muscular tissues from small and large animals (rats, rabbits, chickens, pigs, lambs and beef animals) have been studied immediately post-mortem and also after storage for periods up to several weeks at various temperatures (4°C, 25°C, 38°C). Modifications in the biochemical, structural and organoleptic characteristics of epinephrine treated muscles are shown by a considerable reduction in the exudation of muscular tissue fluids ("drip") and in the structural breakdown of the muscular tissue.

Moreover, the characteristic post-mortem fall in pH of the muscular tissue is inhibited and the organoleptic qualities of meat (and especially tenderness) are maintained or improved.

Combined treatments (epinephrine/irradiation) have given good results with pig meat, beef and poultry meat and, it seems, do not cause the meat to be unfit for human consumption.

* Ce travail a été effectué dans les laboratoires du Battelle Memorial Institut (Genève) et dans les laboratoires de recherches de General Electric (Schenectady).

† Adresse actuelle: Medical School, Vincent Drive, Birmingham 15, England.



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Such a combination process in conjunction with suitable protective packaging materials offers the possibility of better storage properties in the treated meat when held at freezing or atmospheric temperatures.

The question of surface irradiation only or penetrating radiation is bound up with the problem of muscle sterility.

## INTRODUCTION

Dans la pénurie actuelle d'aliments protéiques, la production et la distribution—en fonction des besoins et des disponibilités des différents pays—d'une viande saine et acceptable constitue un des principaux problèmes de nutrition.

La viande déclarée salubre au moment de l'abattage peut être considérée comme une viande pauci-ou même amicrobienne.[1,2]

L'examen clinique "sur pied" et l'application de certaines conditions physiologiques pendant et avant l'abattage (repos, diète hydrique, etc.) ainsi qu'un examen rigoureux anatomo-pathologique et bactériologique post-mortem sont nécessaires pour contrôler la contamination et la toxicité primaires de la viande.

Sa manipulation, dans des conditions "d'hygiène normale" entraîne une contamination secondaire de surface par des micro-organismes saprophytes, "souillure obligatoire", et pathogènes, "souillure d'exception".

Cette viande "hygiénique" est sujette, pendant la période de préconsommation, à une double détérioration:

—l'une d'origine généralement exogène, consécutive à l'activité des micro-organismes,

—l'autre d'origine endogène, due à l'autodétérioration enzymatique. Dans des travaux antérieurs, Zender, Lataste-Dorolle *et al.*[3,4,5] ont étudié les phénomènes d'autolyse aseptique au niveau du muscle.

Pour obtenir une *stabilisation ou une stérilisation des micro-organismes* dans les viandes manipulées, différents procédés physiques et chimiques[6–17] que nous rappelons brièvement, sont utilisés:

—Procédés thermiques: appertisation et cryoconservation (réfrigération, congélation),

—Stérilisation par les radiations ionisantes: électromagnétiques (rayons $\gamma$ et rayons X), et corpusculaires (rayons cathodiques et rayons $\beta$),

—Stabilisation par les antibiotiques.

Ajoutons enfin les procédés de dessiccation et cryodessiccation, la salaison, le boucanage, etc.

Tous ces procédés peuvent être utilisés seuls ou associés.

En ce qui concerne *l'inhibition de l'autodétérioration enzymatique*, peu de "conservateurs" sont employés dans la technologie carnée. L'activité réduite ou la toxicité de ces produits limite leur utilisation.

Récemment, nous avons pu mettre en évidence l'effet anti-autolytique de l'administration ante-mortem de l'adrénaline. Les résultats ont été exposés en détail ailleurs.[16–22]

Dans ce travail, nous nous limiterons à présenter les conséquences sur la conservation de la viande de l'effet anti-autolytique de l'adrénaline, utilisée seule ou en combinaison avec la radiostérilisation de surface (rayons cathodiques, 1,5 MeV).

## ACTION SUR LA CONSERVATION DE LA VIANDE DE L'EFFET ANTI-AUTOLYTIQUE DE L'ADRENALINE

Nous examinons successivement les résultats concernant l'étude, dans des conditions aseptiques et dans des conditions "hygiéniques", de l'effet de l'adrénaline sur la conservation de la viande.

*Conservation de la viande "stérile"**

L'inhibition par l'adrénaline de l'autolyse enzymatique de la viande a été étudiée systématiquement.

Des muscles (longissimus dorsi) d'animaux

*Echantillons de viande prélevés et conservés en conditions aseptiques, n'ayant montré pendant toute la durée de la conservation aucun signe de contamination.

de laboratoire (lapin, rat) et d'abattoir (porc, mouton, boeuf, cheval), ont été

—prélevés et conservés (à l'abri de la lumière) dans des conditions d'asepsie rigoureuse, afin d'éliminer toute contamination microbienne;

—maintenus à des températures de 20° et 37° afin de réaliser les conditions favorables ou optima de l'activité enzymatique,

—et contrôlés immédiatement post-mortem et pendant des conservations de 15, 30, 50 et 150 jours.

Les relations effet/dose et effet/temps ont été mises en évidence par l'administration de doses croissantes (10 à 1000 $\gamma$/kg) d'adrénaline à des temps différents (0 à 24 heures avant l'abattage).

Nous présentons ici les résultats concernant l'étude parallèle des modifications métaboliques, structurales et organoleptiques des muscles d'animaux témoins et d'animaux ayant reçu 250 $\gamma$/kg d'adrénaline quatre heures avant l'abattage.

*Viande non adrénalisée* (témoin). Dans une première phase, le mécanisme d'autolyse est défini par la prédominance croissante des phénomènes de la glycolyse.

La glycogénolyse musculaire avec formation d'acide lactique s'accompagne d'une chute du pH (7,2 à 5,5) et d'une baisse du taux d'ATP, aboutissant à l'établissement du rigor mortis.

L'acidification du muscle entraînerait une modification des propriétés physico-chimiques des protéines musculaires (baisse de solubilité au pH) qui serait à l'origine des phénomènes exsudatifs. Cet exsudat est riche en protéines et en $K^+$ et peut atteindre, en 15 jours, 15% (lapin) à 25% (porc, mouton) du poids du muscle.

La seconde phase du stockage est dominée par l'activité protéolytique qui conduit d'abord à l'attendrissement de la viande, puis à sa détérioration. L'activation des enzymes protéolytiques endocellulaires (cathepsines) serait liée particulièrement à la baisse du pH. Après 15 jours de conservation à 37°, l'examen de la bande électrophorétique montre l'apparition de nouvelles fractions protidiques, plus rapides.

A ces différents stades d'autolyse graduelle (exsudation, attendrissement, détérioration) correspondent diverses altérations structurales se traduisant par une perte de turgescence progressive de la fibre musculaire, un ramollissement général et un clivage à différents niveaux de l'édifice myofibrillaire.

Il existe donc, en dehors de toute action microbienne, une autodétérioration enzymatique du muscle qui conduit principalement à

—une perte de poids par exsudation,

—un effondrement de la structure myofibrillaire,

—et à un avilissement des caractères organoleptiques ou culinaires (viande décolorée, flasque, d'odeur et de saveur désagréables).

Ces résultats confirment les études effectuées par Bate-Smith et Bendall,[23,24] Lawrie,[25,26] Partmann,[27] Sharp,[28] et Wismer-Pedersen[29] sur les modifications métaboliques du rigor mortis, particulièrement les relations entre la glycogénolyse, le niveau de l'ATP, de la phosphocréatine et la baisse du pH.

*Viande adrénalisée.* Sur le *plan métabolique*, l'administration ante-mortem de l'adrénaline entraîne une inhibition indirecte de l'autolyse en modifiant dans le post-mortem certains facteurs de la cinétique enzymatique musculaire.

En effet, elle induit *in vivo* une baisse du glycogène musculaire (réactivation de la phosphorylase inactive) avec formation d'acide lactique, qui entre aussitôt dans le cycle Cori.

Le substrat glucidique étant diminué au moment de l'abattage, l'activité post-mortem des enzymes glycolytiques n'engendre qu'une faible quantité d'acide lactique; le pH du muscle n'est que légèrement abaissé (7,2 à environ 6,7).

Ce maintien du pH au voisinage de la neutralité conserverait les propriétés lyophiliques des protéines (blocage de l'exsudation) et inhiberait la vitesse de réaction des cathepsines pendant la phase de conservation.

Nous présentons dans le Tableau 1, à titre d'exemple, quelques données métaboliques,

TABLEAU 1

| | Muscle témoin | Muscle adrénalisé |
|---|---|---|
| Teneur en glycogène au moment de l'abattage (mg/100 g de muscle) | 4,2 | 0,24 |
| Teneur en acide lactique au moment de l'abattage (mg/100 g de muscle) | 1,75 | 0,55 |
| pH 8 heures après l'abattage | 5,5 | 6,8 |
| Exsudat (ml) après une conservation de 15 jours à 38° | 15,0 | 1,0 |

obtenues chez le lapin ayant reçu 250 $\gamma$/kg d'adrénaline s.c., quatre heures avant l'abattage.

*L'examen morphologique* en contraste de phase montre que le rigor mortis n'est pas apparent. De plus, pendant la conservation, le muscle des animaux adrénalisés garde macroscopiquement et microscopiquement une structure de viande fraîche. Les fibres musculaires sont encore turgescentes et intactes par opposition à celles des muscles non adrénalisés, désintégrées.

En ce qui concerne *les caractères organoleptiques* de la viande adrénalisée, on constate un attendrissement immédiat permettant de supprimer le "ageing" et les autres méthodes physicochimiques d'attendrissement.

L'adrénaline se comporterait comme un "plasticizer". Cet effet pourrait être lié à l'interaction adrénaline/système ATP.[30]

Une stabilisation des caractères organoleptiques (couleur, saveur et tendreté) rapprochant la viande adrénalisée de la viande fraîche, a également été constatée pendant le stockage.

Le mécanisme de l'effet anti-autolytique de l'adrénaline a pu être mis en évidence d'une façon indirecte par les travaux de S. RADOUCO-THOMAS[30,31] lors de l'étude du système adrénergique du muscle.

*Conservation de la viande "hygiénique"**

L'adrénaline s'est montrée un fort inhibiteur des phénomènes d'autolyse enzymatique de la viande, mais elle ne contrecarre pas la détérioration due à la contamination par les micro-organismes.

Les viandes de boucherie adrénalisées, manipulées et donc "souillées" représentent, comme la viande des animaux non adrénalisés, un milieu de culture propice et peut-être plus favorable (pH voisin de la neutralité) à la prolifération microbienne.

Il est donc nécessaire d'associer à l'administration d'adrénaline un des procédés de stérilisation ou stabilisation microbienne signalés: radiations ionisantes, antibiotiques, techniques thermiques, etc.

Nous exposons ci-dessous les résultats obtenus par l'association de l'administration ante-mortem d'adrénaline avec la stérilisation de surface par un flux d'électrons rapides.

## CONSERVATION DE LA VIANDE PAR LE PROCEDE COMBINANT L'EFFET ANTI-AUTOLYTIQUE DE L'ADRENALINE AVEC LA RADIOSTERILISATION DE SURFACE

L'irradiation des échantillons a été effectuée dans les laboratoires de recherche de General Electric avec des rayons cathodiques 1,5 MeV d'un transformateur résonnant, à des doses de 0,50, 0,67, —2, —3, —4, —6 Mrads, à une vitesse de 33.000 rads/sec.

*Conservation de la viande* "stérile" *irradiée*

Les expériences ont été effectuées dans les conditions d'asepsie exposées plus haut afin de mettre en évidence:

—pour la viande non adrénalisée-irradiée,[5] les effets de l'irradiation sur les phénomènes d'autolyse enzymatique.

—pour la viande adrénalisée-irradiée,[20] les relations de l'irradiation et de l'adrénaline (synergie ou antagonisme) sur le plan anti-autolytique.

Les échantillons de viande (lapin, porc,

*Viande prélevée sur des animaux sains et ensuite manipulée.

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boeuf) ont été prélevés aseptiquement, soumis à un flux d'électrons rapides de 0,5 à 4 Mrads et conservés dans les mêmes conditions que les échantillons non irradiés pendant 15 jours à 37°C.

Dans les *viandes non adrénalisées-irradiées*, la chute post-mortem du pH n'est que très faiblement inhibée, l'effet des rayons cathodiques sur les enzymes de la glycogénolyse semble donc minime. Il pourrait s'agir d'une légère inhibition des enzymes à —SH du catabolisme glucidique dont la sensibilité de structure aux irradiations *in vitro* est toutefois beaucoup réduite *in situ*.

Doty *et al.*[33] ont obtenu des résultats similaires avec des radiations gamma: des doses de 8 Mrads n'ont entraîné qu'une inhibition de 0,3 à 0,5 unités de pH.

Dans la période qui suit le rigor mortis, l'activité des enzymes de la protéolyse, favorisée par la chute du pH, ne semble guère inhibée par les irradiations.

Signalons cependant qu'une inhibition significative de la désintégration structurale a pu être mise en évidence par l'étude parallèle de minces coupes superficielles et profondes.

Ces données suggèrent que l'irradiation de surface n'inhiberait que peu ou pas efficacement les phénomènes de l'autolyse enzymatique de la viande et se montre insuffisante pour assurer sa conservation en milieu aseptique.

En ce qui concerne les caractères organoleptiques de la viande, non seulement l'irradiation ne supprime pas leur dépréciation par l'autolyse, mais elle y surajoute encore, aux fortes doses, des modifications de couleur et de palatabilité liées à ses effets secondaires sur les constituants de la viande.[31–36]

*Viandes adrénalisées et irradiées*. L'étude des modifications du métabolisme a permis de mettre en évidence que l'effet anti-autolytique de l'adrénaline n'est pas modifié dans les échantillons ayant subi une irradiation de surface. L'inhibition de la chute du pH, de l'exsudation et de la protéolyse sont du même ordre de grandeur que dans les muscles adrénalisés non irradiés. Toutefois les fortes doses (4 Mrads) semblent diminuer l'action anti-exsudative de l'adrénaline.

L'étude des caractères structuraux des fibres et myofibrilles n'a pas révélé de différences significatives entre les zones de surface et de profondeur.

En ce qui concerne les caractères organoleptiques, l'amélioration de la tendreté due à l'adrénaline se retrouve dans les échantillons soumis à l'irradiation, mais certains effets secondaires de l'irradiation apparaîtraient pour les fortes doses.

Ces résultats montrent que, quelle que soit la dose, l'irradiation de surface ne présente qu'un effet anti-autolytique insignifiant.

L'adrénaline reste la seule responsable de l'inhibition de l'autolyse enzymatique pendant la période de conservation consécutive à l'irradiation.

*Conservation de la viande "hygiénique" irradiée*

Nous présentons quelques résultats préliminaires concernant l'effet de la radiostérilisation de surface sur la conservation et sur les caractères organoleptiques des viandes adrénalisées et manipulées dans des conditions hygiéniques.

Des échantillons de viande de *porc*, adrénalisée et non adrénalisée, ont été

—prélevés dans les conditions hygiéniques d'abattoir,

—irradiés avec des doses de 0,67 à 6,6 Mrads,

—puis conservés dans des sachets de polyéthylène à des températures normales (20°) et tropicales (37°).

Les résultats suivants ont été enregistrés: *pour les viandes conservées* à 20°,

—les échantillons *non adrénalisés* et soumis à des radiations de 0,67 et 2 Mrads étaient détériorés après 8 jours, ceux soumis à des irradiations de 3 et 6 Mrads étaient détériorés après 19 jours,

—les échantillons *adrénalisés* étaient tous satisfaisants après 19 jours, sauf l'échantillon non irradié.

La dose minima de 0,67 Mrad serait donc suffisante pour conserver pendant 19 jours des échantillons adrénalisés, alors qu'une dose de 6 Mrads serait insuffisante pour conserver, dans les mêmes conditions, des échantillons non adrénalisés.

*pour les viandes conservées à* 37°,

—les échantillons non adrénalisés et irradiés (0,65 à 6 Mrads) ainsi que l'échantillon adrénalisé mais non irradié, étaient altérés après 4 jours, alors que tous les échantillons adrénalisés et irradiés (même avec 0,65 Mrad) étaient bien conservés.

De même pour la viande de *poulet*. Les échantillons témoins, irradiés avec 0 à 6 Mrads, étaient détériorés après 5 jours à 37° alors que, dans les mêmes conditions, les échantillons adrénalisés et irradiés avec 3 et 6 Mrads étaient satisfaisants.

Après 12 jours à 20°, les échantillons non adrénalisés, irradiés avec 0,67–2–3 Mrads, étaient détériorés. Les échantillons irradiés avec 6 Mrads s'étaient conservés mais présentaient la forte odeur caractéristique des viandes irradiées. Les échantillons adrénalisés et irradiés avec 3 et 6 Mrads étaient bien conservés, et, en outre, dépourvus d'odeur désagréable.

Des résultats similaires ont été obtenus avec la viande de jeune boeuf (longissimus dorsi) et de poulet (pectoraux).

Des essais préliminaires, suggérant eux aussi une éventuelle synergie adrénaline/irradiation, ont été effectués avec la viande de *jeune boeuf*.

Au vu de ces résultats, on peut se demander si, outre son effet anti-autolytique, l'adrénaline ne pourrait pas permettre une utilisation plus fructueuse des irradiations.

Il est possible que la stabilisation des réactions métaboliques du post-mortem qu' engendre l'adrénaline, puisse permettre de choisir des conditions d'irradiations et de post-irradiation ($t°$, $O_2$) engendrant soit un meilleur effet bactéricide des radiations ionisantes, soit une diminution de leurs effets secondaires.

De plus, l'adrénaline, en agissant *in vivo* sur le métabolisme des graisses, semble diminuer la dépréciation des caractères organoleptiques de la viande, due aux fortes doses d'irradiation.

Il serait donc possible d'envisager favorablement l'association adrénaline/radiations ionisantes:

—d'une part, l'adrénaline contrecarre l'autolyse enzymatique sur laquelle les irradiations n'agissent que faiblement,

—d'autre part, elle pourrait, éventuellement, dans certaines conditions, favoriser leur effet anti-microbien et antagoniser leurs effets secondaires.

Le choix entre une irradiation de surface ou une irradiation pénétrante reste lié au postulat de la "stérilité" du muscle à l'abattage, problème encore fort discuté.

Jepsen[37] et Fredholm,[38] admettent que les viandes provenant de "healthy and physiologically normal animals are sterile". Legroux et Jéramec,[39] Verge,[40] Lepovetsky,[41] Jensen,[42] Keller,[43], Dubos,[44] Cosnett *et al.*[45] et Drieux[46] rappellent la possibilité d'une contamination du muscle en profondeur chez les animaux déclarés sains. Olivant[47] présente une série de résultats concernant l'examen bactériologique de tissus d'animaux "normaux" et conclut que des bactériémies transitoires, liées aux stress physiologiques avant et pendant l'abattage, pourraient être à l'origine d'une contamination en profondeur du muscle. Ingram[48] estime que "samples of muscles removed aseptically from normal healthy animals have proved to contain small numbers of bacteria in a large proportion of cases".

Un travail systématique sur des tissus d'animaux "normaux", effectué dans des "conditions de laboratoire et d'abattoir", serait nécessaire pour décider de l'opportunité d'une radiostérilisation agissant en surface ou en profondeur.

L'utilisation de l'adrénaline comme anti-autolytique[49] en liaison étroite avec d'autres techniques anti-microbiennes adéquates (antibiotiques, processus thermiques, etc.) et un emballage protecteur convenable, est également envisagée.

*En résumé*, le procédé combiné d'inhibition des phénomènes autolytiques par l'adrénaline et de stabilisation-stérilisation des micro-organismes par l'irradiation pourrait permettre une meilleure et plus longue conservation de la viande et de ses produits de transformation, aux températures de congélation, normales et même tropicales, et pourrait assurer ainsi une répartition plus

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efficiente de la viande entre les différents pays.

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# The Present Status of Irradiation of Foods in the U.S.A.—Meat, Poultry and Fish

B. S. SCHWEIGERT

American Meat Institute Foundation and Department of Biochemistry,
The University of Chicago, Chicago, Illinois, U.S.A.

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THE greatest attention has been focused in developing satisfactory irradiated foods from this group of foods because of their importance in the human diet and because of their relative cost per unit weight. Some years ago, it was considered that meat and milk were the most sensitive to irradiation treatment in terms of the development of off-flavors and odors. Recent studies indicate, however, that selected applications of irradiation may show promise for certain food items in these groups.

Meats treated with sterilization dosages show an increase in the amounts of methyl mercaptan, hydrogen sulfide and carbonyl compounds produced, and a rise in pH values. A decrease in the amount of the sulfur containing tripeptide, glutathione, and of glycogen was also observed. These chemical studies, which have been designed to ascertain the chemical changes that occur in foods during irradiation, have now been correlated with taste panel evaluations in collaborative studies that we have carried out with Dr. A. M. Pearson at Michigan State University. Although either hydrogen sulfide, methyl mercaptan, or carbonyl compounds was a fair indicator of panel scores, correlation coefficients for hydrogen sulfide $\times$ methyl mercaptan $\times$ carbonyl compounds with panel scores gave correlations of 0·93, 0·65 and 0·61 for beef, pork and veal respectively. Thus, considerable progress has been made in ascertaining the nature of off-flavor and odor constituents. Unfortunately, no simple technique to prevent the occurrence of these off-flavors and odors has as yet been devised. Several factors appear to be helpful however. These include irradiation at low temperatures, keeping oxygen away during and after irradiation, inactivation of proteolytic enzymes by heat prior to irradiation, and irradiating meats with a relatively high fat content. This latter observation suggests that the major off-flavor constituents arise from components of the lean portion of meat. One can mask these undesirable flavor changes by the addition of additives or by dilution. Acceptability studies with Army personnel have shown a high acceptability, for example, of pre-cooked pork roasts that were irradiated and stored prior to final preparation and serving. Thus, we should not lose sight of the importance of consumer evaluation as compared to critical results obtained with analytical taste panels which are trained to detect small differences.

On an over-all basis, pork products show much less off-flavor change than do beef products. Irradiated chicken also appears to be quite acceptable. Irradiated pre-cooked chicken shows an improved flavor on high temperature storage, perhaps to a greater degree than is observed for other meats.

In addition to the promise shown for the use of sterilization dosages for certain pork products and chicken, the possibilities for the use of low level irradiated dosages to prolong the shelf life of prepackaged meats appear promising. The use of 50,000 to 100,000 rads treatment has been shown to prolong the shelf life of prepackaged meats two to five-fold when kept under refrigeration. These techniques are quite effective in

reducing the number of pseudomonas organisms which are radiation sensitive and normally spoil meat at refrigeration temperatures.

The possibilities of low level dosages prolonging the shelf life of poultry and fish are equally promising. Fish appear to be more sensitive to the development of rancidity and color changes because of the greater susceptibility of fish oils to oxidation. Codfish cakes show a rather high acceptability, even after treatment with sterilization dosages. This is also true for other sea food products, such as cooked crab meat and oysters. The carotenoids associated with the normal pink color of salmon or shrimp may turn brown or be destroyed by sterilization dosages which make the products undesirable from an appearance standpoint.

On an over-all basis, therefore, irradiation preservation of meats, poultry and fish appears promising for certain products and certain selected applications. Undesirable flavor and odor development is the major factor in the acceptability of other irradiated products. It will require a major scientific "break through" to circumvent these serious flavor and odor problems before one could anticipate wide acceptance of products such as fresh beef treated with sterilization dosages in the United States. If we consider techniques for preparing combination dishes, masking the flavor changes with gravies, spices, etc., some broader market than previously indicated might be possible.

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# The Present Status of Radiation Preservation of Meat and Meat Products

E. STENBERG KNUDSEN

The Danish Meat Research Institute, Roskilde, Denmark

A brief account on the current ways of meat preservation is given in connexion with an evaluation of radiation preservation of meat. A conclusion is that it would not be feasible to replace the current preservation methods by irradiation. But there seems to be some advantage in combining radiation treatment with cooling to prolong the storage life of the products. And in the same way it seems to be feasible to combine the ordinary heat preservation with a radiation preservation to achieve a meat-preserve with good keeping quality and improved texture.

The possibility of applying radiation as a preservation technique to prepacked meat products is mentioned.

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THE possibility of using ionizing radiations in the preservation of foodstuffs received little attention in Denmark and perhaps also in many other European countries until 1950 when Heen published his work carried out in 1949 in the U.S.A. with a Van de Graaff accelerator and until Professor Proctor and his group at M.I.T. in the United States began publishing their extensive work on the subject. Later information became available concerning the very extensive activities of the Quartermaster Food and Container Institute of the U.S. Armed Forces. In addition, two excellent review papers which appeared in 1955 by Professor J. Kuprianoff from Bundesforschungsanstalt für Lebensmittelfrischhaltung, Karlsruhe, and the comprehensive review by Dr. R. S. Hannan from the Low Temperature Research Station in Cambridge should be mentioned. This information awakened other research workers to the possibilities of using this new technique in helping to solve problems in the preservation of meat and meat products.

It is necessary to consider the kind of preservation problems involved in the marketing of meat and meat products and to determine whether in the light of present knowledge of food irradiation, it is likely that any of these problems can be solved by this new process of food treatment.

In dealing with these questions it must be mentioned that the need for preservation varies considerably according to conditions of climate, methods of distribution, state of technical development, consumer density, etc. Moreover the difference in keeping quality of different types of meat must be borne in mind. It may be advisable to divide the problems into two main groups, namely those problems concerned with bulk preservation of meat for manufacturing or wholesale use, and those concerned with consumer-size cuts of meat and meat products, either unwrapped or packed in cans, foils, paper, etc.

Without attempting to survey the world distribution of meat, it would be useful to consider some aspects of the preservation of bulk cuts of beef and pork for transport and distribution over long distances.

Beef is stored and distributed mainly in the chilled or frozen form and the storage life of chilled meat varies from 1 to 4 weeks according to sanitary conditions and storage temperature (0° to 4°C). The question arises whether the use of ionizing radiations will displace refrigeration for the transport of meat over long distances. This is unlikely

to happen for, although irradiation may inactivate micro-organisms in meat, with the range of dosages at present being considered, it does not prevent enzymatic deterioration which occurs when meat is stored at normal atmospheric temperatures. Moreover there is the danger of recontamination which would be more serious at room temperature storage than during cooler storage because of the more rapid multiplication of micro-organisms at these higher temperatures. For these reasons it seems necessary to continue to store and distribute fresh beef at chilling temperatures.

There is the further question whether irradiation will prolong the storage life of chilled fresh meat. Much investigational work has been carried out on this subject and it has been shown that irradiation gives rise to off-flavours and off-odours which, in the case of beef, are rather serious. Since this effect is increased by increasing radiation doses, the applied doses must be kept below a certain threshold. It appears that, with the application of a low dose of about 100,000 rads the increase of storage life under refrigeration is between two and five-fold, and this dose seems to be slightly below the threshold for detection of flavour change. However, some bacteria which are able to cause spoilage in refrigerated meat have a much higher radiation resistance than ordinary spoilage bacteria. The preservative effect of this relatively low dosage may be somewhat limited and the use of an antibiotic treatment or other additives to supplement the effects of irradiation has therefore been suggested. If more extensive tests show that this combination of irradiation with chilling comes up to expectations, then it may replace freezing for many purposes, since freezing is a rather unsatisfactory method for bulk cuts of beef, because of the "drip" which occurs when the meat is thawed.

In the case of pork the situation is somewhat different because pork is less sensitive to irradiation since it can stand a higher dose without developing adverse flavour changes than, for example, beef and poultry. It is evident that a dose of about 1 Mrad could be applied to pork without serious changes and there is some possibility that the storage life of chilled pork could be prolonged appreciably by a combination of cooling and radiation.

A considerable part of pork is distributed as bacon and other cured products and the keeping quality of such products varies according to the salt content and the conditions of storage and distribution. The storage life of bacon having a salt content of 3–4 per cent is about 1–2 weeks at 10°C and about 3 months at −3°C. In the latter case the storage life is mainly determined by the extent of development of rancidity in the fat. With irradiation it would seem to be possible to prolong the storage life of bacon as regards microbiological spoilage but the problem of rancidity tends to become even more serious when the product is irradiated. Therefore at present it seems to be necessary to carry out the irradiation in an oxygen-free atmosphere, which means that it should be done either *in vacuo* or in an inert gas. Storage before irradiation should be under the same conditions. The most obvious application would probably be for mild cured prepacked bacon, sausages and similar products as mentioned below.

Owing to the rather limited sizes of the radiation sources, all the experiments on meat preservation by ionizing radiations which have been carried out so far have been with rather small samples. Future experiments on the pilot plant scale will presumably give clearer answers to the question of the feasibility of using radiation in the preservation of bulk cuts of meat.

Turning now to consumer-size cuts of meat which include a variety of meat products, it must be emphasized that these have a rather complex set of characteristics, determined by the nature of the raw material and by the different methods used in processing and treatment. This means that the radiation treatment of every product has to be carefully investigated in order to prove or disprove the advantage of irradiation for each individual product.

In considering some methods currently

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used for meat preservation such as drying, curing, heat treatment, cooling and freezing, the question is whether the use of irradiation is likely to replace any of these treatments or whether it could be used in combination with them to improve the quality of preserved meat products.

As an example, the heat treatments of meat packaged in a tin can could be considered. Owing to the poor heat conductivity of the product, the heat processing of canned foods is fundamentally a rather poor method of preservation since the sterilizing value of the heat process has to be judged by the treatment (temperature and heating time) received by the contents at the centre of the can where the chance of survival of bacterial spores is greatest. This means that the outer layers of the product have to be overprocessed and this leads to a decrease in quality and to unnecessary shrinkage. This is especially the case when the sterilizing requirement is high and a severe heat treatment has to be applied. This raises the question whether irradiation could replace heat treatment in the canning process. At present this does not seem to be feasible because of the adverse side effects produced by the high doses necessary for destroying enzyme activity and toxin-producing micro-organisms. However, it has been shown that a combination of heat and irradiation may prove to be beneficial. A few examples may be quoted: For beef it has been found that in canned samples precooked to about 80°C and then irradiated with about 2 Mrads, the quality after storing at room temperature for 8 months was still good. Hams heated to 67°C and then irradiated with a dose of 0·2 Mrad showed no significant adverse effects and there were indications that this treatment was sufficient to produce a commercially sterile product. (This, however, is probably true only for hams containing more than the usual quantity of salt.) Pork sausages have been irradiated with 2 Mrads without adverse effects. In other experiments samples of pork luncheon meat were heated to maximum internal temperatures of 65, 75, 85 and 95°C and afterwards irradiated with 0·5, 1 and 2 Mrads. No adverse effects on colour, taste or odour were found with radiation doses of 0·5 and 1 Mrad, but a dose of 2 Mrads significantly decreased the organoleptic quality of the products. The samples were stored at 30°C, and after 9 weeks the numbers of spores or spore-forming bacteria in the samples irradiated with 1 and 2 Mrads was still very low. By contrast multiplication of this type of bacteria had taken place in the control and in the sample irradiated at 0·5 Mrad. The total number of aerobic bacteria after 9 weeks, in samples irradiated with 1 and 2 Mrads and heated to 85 and 95°C was very low and these samples could be regarded as commercially sterile. Larger numbers of bacteria were found in the sample treated with lower doses and at lower temperature.

Many types of canned meat products are semi-perishable and must be stored under refrigeration. If irradiation could replace refrigerated storage it would mean a vast extension of the market for such products.

The production and sale of various types of foods prepackaged in different types of plastic films is increasing rapidly and these foods include meat and meat products. The storage life of meat products packages in films depends on various factors, particularly the characteristics of the product itself, but the quality of the packaging film including the seal, the packaging technique and the storage temperature are also important. In many cases it is found that the storage life of such packaged products is rather short, especially at room temperature, but also at lower temperatures.

This is a field where it may be possible to apply moderate doses of radiation as a preservation treatment. The results of an experiment with irradiation of sliced bacon packaged in film may be quoted. The bacon was of the Wiltshire type with a salt content of about 3 per cent (water about 30 per cent, salt in water average 10 per cent). The film was a laminate of polyethylene and cellophane and the bags were vacuum-sealed. Samples irradiated with a dose of 1 Mrad (electrons) and stored 6 months at 5°C were rated as acceptable by a very experienced and critical taste panel and the number of

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living micro-organisms were decreased by a factor of about 1 million compared with the unirradiated control. Irradiated samples stored at 20°C for 2 months were likewise rated as acceptable and the number of living micro-organisms was decreased by a factor of 10,000.

There are still many contradictions in the experimental results achieved in the field of radiation preservation. In some cases this may be due to the different characteristics of products known by the same name but differing appreciably in composition according to the place where they are produced. In other cases lack of confirmatory experiments and of information concerning experimental conditions, etc. are probably responsible for some of the existing confusion. It would seem that much investigation is still needed before it can be established clearly which products give the best results when treated with ionizing radiations. This applies to the effect of irradiation on spoilage micro-organisms under different conditions as well as to the effect on food poisoning organisms, especially in products which are stored for long periods without refrigeration.

In the present paper it is still assumed that the products are to be supplied to the civilian market and further that the consumers are highly discriminating as is the case for example in Western Europe and the U.S.A. The picture may change somewhat in the case of food to be preserved for consumption by troops, or if irradiated meat products are to be marketed in areas where people may be prepared to accept foods in which the flavour and appearance have been somewhat altered. In such cases the chief requirement would be the wholesomeness and nutritive value of the food.

Finally a summary will be given of plans in Denmark for the future investigation of radiation preservation of meat. It is unlikely that experiments with full sterilizing doses (about 5 Mrads) will receive much attention, but technological experiments will be made with the purpose of investigating the influence and interaction between radiation, heating, cooling and curing. Research on the value of free radical acceptors in the process and microbiological research to identify the types and properties of micro-organisms that survive and multiply in different irradiated products will also be undertaken. A prerequisite for carrying out these experiments is, of course, the availability of radiation sources of $\gamma$- and electron rays and these are or will be available at the Danish Atomic Energy Establishment.

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# The Present Status of Radiation Preservation of Food Products—Fish

J. M. SHEWAN

Torry Research Station, Department of Scientific and Industrial Research
Torry, Aberdeen

The sterilization of the commoner varieties of sea foods by irradiation requires doses which almost invariably produce undesirable and unacceptable organoleptic changes in these products. It seems unlikely, therefore, that this method can at present compete commercially with the more conventional preservation processes. Recent work, particularly in the U.S.A., suggests that pasteurizing doses either alone or along with a variety of "combination" treatments, could give satisfactory products with greatly enhanced shelf life particularly at chill temperatures. Of over 20 species of fish tested, cod and halibut stand up best organoleptically. In fatty fish, such as herring, rancidity of the oil is an added hazard, while in other species like salmon, the pigment is bleached or in white fleshed species, brown discolorations appear. On the other hand, blanched oysters and cooked crab and shrimp meats stand up well with great improvements in their keeping qualities. Provided the more obvious defects in the present process can be overcome, it seems likely that this new technique could revolutionize the present methods of marketing unfrozen fish to inland populations far from the ports.

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It is well recognized that fish and fishery products are extremely perishable and it is not surprising, therefore, that they were among the first materials to be subjected experimentally to irradiation. It has become obvious from the later work of Brasch and Huber[1,2] and of Proctor *et al.*[3,4] at the Massachusetts Institute of Technology that the potentialities of irradiation as a preservative method are by no means so spectacular as was at first thought. This is due mainly to the fact that the dosage required for sterilization, which was the first objective of the early work, produces changes in the odour or flavour of the products that render them quite unacceptable to most consumers.[5]

Subsequent work has shown, however, that some species of fish stand up better to irradiation than others and that these when subjected to only pasteurizing levels suffer none of the undesirable changes produced by sterilization. As a result most attention is now being directed to pasteurization, usually along with "combination-treatments" such as blanching, addition of antibiotics or chemical protectors with a view to improving both the shelf life and the organoleptic properties of the irradiated materials. The bulk of the present work in this field is now being done in U.S.A. by the Fish and Wildlife Service, in collaboration with Institutes such as the Massachusetts Institute of Technology, Oregon State College and Maryland State University and industrial laboratories.[6,7] Much of this work forms part of the U.S.A. defence programme; but sufficient material has already been released to afford an overall picture of progress made.

## STERILIZATION

As already mentioned little work is now being done on the sterilization of fish and fishery products. Experience from a variety of sources has indicated that fish requires a dose in the region of 2 Mrads to produce a commercially sterile product[5] and a dose of 4·8 Mrads to produce one free from *Cl. botulinum* spores. As will be seen from Table 1,

TABLE 1. The maximum permissible irradiation doses that can be given to various fish and fishery products (from various sources)

| Species of fish | Irradiation dose* (Mrad) |
|---|---|
| Butterfish | 0·50–0·70 |
| Butterfish (blanched 140°F for 5 min) | 1·5 |
| Cod | 0·8–1·5 |
| Cod (blanched 140°F for 5 min) | 1·5 |
| Haddock | 0·60 |
| Halibut | 1·0–2·0 |
| Lemon sole | 0·5–1·0 |
| Ling | 0·5–1·0 |
| Mackerel | 1·0 |
| Pollack | 0·5–0·70 |
| Pollack (blanched 140°F for 5 min) | 0·80 |
| Salmon | 0·5 |
| Tuna | 1·0 |
| Whiting | 0·20–0·25 |
| Whiting (blanched 140°F for 5 min) | 0·80 |
| Ocean perch | 0·80 |
| Kippered herring | 1·0 |
| Kippered Sable fish | 1·0 |
| Kippered Sturgeon | 1·0 |
| Crab meat | 1·0 |
| Shrimp | 1·0 |
| Pacific Oysters (blanched) | 5·0 |

* Doses in excess of those given above produce undesirable organoleptic changes, e.g. in flavour and odour and/or in appearance, e.g. bleaching of pigment in salmon.

these doses are usually in excess of those that produce undesirable and unacceptable organoleptic changes in the commoner species of fish. The undesirable odours and flavours produced, which have been variously described as "metallic", "burnt-feather like" and "rubbery", can, of course, be detected after irradiation at levels much lower than those given in Table 1.

In addition to the defects just mentioned, which are immediate results of irradiation, there develop during storage strong bitter flavours, brown and other discolorations of the flesh (particularly noticeable in white-fleshed species), and a toughening of the texture. These changes are all accelerated as the storage temperature is raised, and in fact the whole picture is somewhat reminiscent of that associated with the storage of dehydrated fish.[8] If, as is generally believed, irradiated fish must be of exceptionally high quality if it is to compete successfully with other preserved products then it seems logical to conclude that irradiation sufficient to effect sterilization is not likely to be a commercial proposition.

In passing it is of interest to note that the sterilization technique has been used in research with a view to identifying the specific contributions made by autolysis and bacterial activity to fish spoilage. As a result of such work PROCTOR *et al.* have concluded that in haddock[9] autolysis plays little part in spoilage during storage in ice, whereas in mackerel[10] enzymic action can be quite considerable.

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## PASTEURIZATION

Whilst sterilization appears, therefore, to be of little more than academic interest, the possibility of developing a satisfactory technique of pasteurization cannot be so dismissed, and as already mentioned, most of the work now in progress both here and in the U.S.A. is being devoted to further examination of this possibility. Here the main feature is the remarkable extension of shelf life obtained when the product is held at chill temperatures (0–5°C). Although the search for the species of fish and shellfish most suitable for pasteurization is still continuing, it will be evident from Table 1 that of those already tested, halibut and cod suffer least organoleptic change; and indeed most of the work on pasteurization has, in fact, been done on the latter species.

Data concerning the extension of shelf life of various products are given in Table 2. It will be noted that with cod fillets irradiated at levels of from 0·25 to 0·50 × Mrad the limit of edibility can be extended by from about 20 to 100 days at 0°C and by about 30 to 50 days at 5°C. With pollack and butterfish the extension is of the order of 10 to 20 days at 1·5°C and with crab meat 50 days at 0°C.

As might be expected these extensions of the shelf life are due, in a large measure, to

TABLE 2. Extension of shelf-life of fish and fishery products (from various sources)

| Species | Irradiation treatment | Extension of shelf-life in days over the untreated controls |
|---|---|---|
| Cod, pollack, butterfish | Blanched, vacuum packed in tins, at 0·5 Mrad | 20 days stored at 1·5°C |
| Pollack and butterfish | Raw, vacuum packed in tins at 0·25 Mrad | 10 –20 days stored at 1·5°C |
| Cod | Raw, vacuum packed in tins at 0·25 Mrad | About 70 days stored at 1·5°C |
| Cod | As above at 0·25 Mrad | About 40 days stored at 0°C |
| | As above at 0·50 Mrad | About 40–50 days stored at 5°C |
| | As above at 0·75 Mrad | About 50–100 days stored at 0°C |
| Pacific cod fillets | Raw, in polythene, at 0·20 Mrad | About 40 days stored at 1·5°C |
| Haddock fillets | Raw, in cellophane, at 0·6 Mrad | About 30 days stored at 1 to 4°C |
| Pacific cod fillets | Raw, at 0·25 Mrad | About 40 days stored at 0°C |
| | Raw, at 0·75 Mrad | About 100 days stored at 0°C |
| Cod fillets | Raw, in polythene at 0·25 Mrad | About 20 days stored at 0°C |
| Crab meat | Raw, at 0·50 Mrad | About 50 days stored at 0°C |
| Crab meat | Cooked, at 0·40 to 0·85 Mrad | About 40 days stored at 0°C |

the reductions in the bacterial load as a result of irradiation (Table 3). However, detailed bacteriological analysis of the flora of the fish before and after irradiation and during storage, has shown that pasteurization not only reduces the bacterial load but also has a marked selective action on the various microbial groups present. In particular the pseudomonds, which are known to be very active in fish spoilage,[12] are almost completely eliminated. This is particularly evident from Table 4, which gives the results of our own experiments with cod.[11] It will be noted that after irradiation the percentage of *Pseudomonas spp.* has fallen from 40 in the controls to about 5, and that the main group now present is the *Achromobacter*. It will also be observed that dipping for 30 min in a 20 p.p.m. solution of chlortetracycline ("aureomycin") before irradiation itself reduces the relative proportion of *Pseudomonas spp.* and that when this treatment is combined with irradiation, virtually all micro-organisms are eliminated except yeasts.

TABLE 3. Reduction in the bacterial load in fish muscle immediately after irradiation (expressed as Log No. per g of muscle)

| Species of fish | Unirradiated controls | After irradiation doses in Mrad of: | | | | | | |
|---|---|---|---|---|---|---|---|---|
| | | 0·1 | 0·2 | 0·25 | 0·4 | 0·5 | 0·6 | 1·0 |
| Haddock fillets | 5·5 | | | | 2·5 | | 1·75 | |
| Cod fillets | 6·5 | 4·0 | 2·0 | | | | | |
| Cod fillets | 6·3 | | | 2·7 | | 2·0 | | Commercially sterile |

During storage at 0°C, the *Pseudomonas spp.* gradually reassert themselves so that by the twenty-first day they constitute over 90 per cent of the flora in the irradiated fish. In these experiments the fish that were given the combined antibiotic–irradiation treatment, had a very low count at the end of the storage period of 23 days and organoleptically were still considered to have been of reasonably good quality. It seems safe to conclude that this enhanced storage life must be related to the elimination or suppression of the active spoilage types belonging to the *Pseudomonas* group.

The remarkable extension in the shelf life of pasteurized fish and fishery products has already caused a considerable amount of interest, particularly in U.S.A. Provided that some of the more disagreeable odours and flavours produced in the fish themselves

TABLE 4. Qualitative analysis of the bacterial flora of irradiated fish during storage at 0°C

| | Normal untreated fish | | | 0·5 Mrad treated fish | | | 0·25 Mrad treated fish | | | Aureomycin dipped fish | | | | Aureomycin dip and 0·25 Mrad treated fish | | |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| Bacterial flora from plates incubated at 20 and 0°C | 1 day after filleting | 16 days at 0°C | 23 days at 0°C | 1 day after treatment | 13 days at 0°C | 21 days at 0°C | 1 day after treatment | 13 days at 0°C | 21 days at 0°C | 1 day after dipping | 9 days at 0°C | 16 days at 0°C | 23 days at 0°C | 6 days at 0°C after treatment | 13 days at 0°C | 20 days at 0°C |
| Total organisms examined | 100 | 200 | 200 | 40 | 100 | 100 | 35 | 100 | 100 | 75 | 100 | 100 | 100 | 50 | 90 | 100 |
| *Pseudomonas* | 40% | 55% | 72% | 5% | 52% | 100% | 3% | 7% | 99% | 32% | 5% | 7% | 7% | 2% | 89% | 97% |
| *Achromobacter* | 30% | 40% | 13·5% | 40% | 47% | — | 63% | 93% | — | 35% | 16% | 9% | 6% | 2% | 11% | — |
| *Flavobacter* | 11% | 1% | — | 2·5% | — | — | 8% | — | — | 5% | 11% | 4% | 1% | — | — | — |
| *Corynebacterium* | 6% | 2% | 1% | 5·0% | 1% | — | 6% | — | 1% | 12% | 18% | 3% | 8% | — | — | — |
| *Micrococcus* | 1% | — | — | 7·5% | — | — | 20% | — | — | 5% | 38% | 15% | 2% | 2% | — | — |
| Yeasts | — | — | — | 2·5% | — | — | — | — | — | 1% | 2% | 55% | 67% | 92% | — | 1% |
| Miscellaneous | 12% | 2% | 13·5% | 37·5% | — | — | — | — | — | 10% | 10% | 7% | 9% | 2% | — | 2% |

or by the packaging materials used, can be eliminated, say by the "combination-processes" already mentioned or by the more recent "tempering" technique (i.e. storage at temperatures of 45°F and above for a certain time followed by storage near freezing point) and provided that, nutritionally, the irradiated products are quite wholesome,[13,14] then the fishing industry will have at its command a most valuable new method for the short term preservation of its products. In particular, it may help to solve some of the difficulties now being encountered by the trade in the marketing of prepackaged unfrozen fish, e.g. for self-service stores; and it could revolutionize the present methods of marketing fish to inland populations far from the ports in countries like U.S.A. and Canada, by making available to them for the first time unfrozen fish and other fishery products of qualities equal to those now being enjoyed by the consumers at the coast.

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# Discussion

Mr. GREENWOOD (U.K.): The target for adrenaline in the body is the phosphorylase system, and the effect of adrenaline treatment is to convert liver glycogen to glucose, but in the muscle, the glycogen is changed to lactic acid, which should lower the pH, rather than raise it. Have various muscle types been studied in respect to the effect of antemortem injection of adrenaline?

Dr. RADOUCO-THOMAS (Switzerland): Although muscle glycogen is converted to lactic acid by adrenaline treatment this is removed by the bloodstream, *in vivo*. This was shown by our examination of lactacidemia and of muscular glycogen and lactic acid post mortem. Regarding the examination of various types of muscles, our experiments have been made with both laboratory (rats, rabbits) and slaughter-house (pig, mutton) animals. Within the limits of our experience the phenomena produced by adrenaline treatment have been quite general for the muscle types studied.

Dr. LAWRIE (U.K.): Are the effects produced by adrenaline due solely to the resultant changes in pH of the muscle after *rigor mortis*?

Dr. RADOUCO-THOMAS (Switzerland): It seems that the effects produced by adrenaline are highly correlated with the resultant changes in pH of the muscle, but there is probably also a direct or indirect specific action of adrenaline. A dissociation could be suspected in relation to the preservation of meat on the one hand and with the improvement of its organoleptic properties on the other.

Dr. INGRAM (U.K.): The Battelle investigations[1,2] have two aspects: surface irradiation of meat, and subsequent autolytic changes. Though the latter was investigated in connexion with the former, it is essential to distinguish between the two. The work on autolysis is interesting but the proposals for the surface sterilization of meat seem unsound.

These proposals rest on the assumption, which was stated clearly, that "hygienically cut fresh muscle from healthy animals presents no inside bacterial contamination". If that were so, irradiation would be required only to destroy surface contamination. Unfortunately, however, the basic assumption is probably incorrect, at least in relation to normal meat practice.

This appears from recent investigations made by competent workers independently in different countries. Samples of muscle removed aseptically from the carcasses of "normal healthy animals", have proved to contain small numbers of bacteria in a large proportion of cases. The data are summarized in Table 1.

The general picture displayed in the table confirms various earlier observations (e.g. references 5, 6).

It is striking that the experience of the

TABLE 1. The proportions which contained bacteria, of aseptically excised muscle samples from carcasses of normal healthy animals

| Investigators | Wt. of sample (g) | No. of samples contaminated/No. examined | | |
|---|---|---|---|---|
| | | Rabbit | Pork | Beef |
| OLIVANT[3] | 1–2 | 0/5<br>0/20 | — | 15/100 |
| PANTALEON[4] | ca. 50 | — | 28/62 | 129/205 summer |
| ZENDER *et al.*[1] | ca. 20 | 1/? | — | — |
| RADOUCO-THOMAS *et al.*[2] | | — | 0·5% | 25% |

Battelle workers (mentioned in their Geneva papers) tallies with that of others; it is significant that the proportion of contaminated samples appears to be unusually low for rabbits, with which the Battelle group have mostly worked, and that the proportion is so high for beef that every sample weighing more than about 100 g seems likely to contain bacteria. A large proportion of these bacteria moreover, are streptococci coliforms and clostrida which appear probably faecal in origin, though this question needs much further investigation.

Plainly, in this situation, surface sterilization would be a highly unreliable and even dangerous process, if applied to meat prepared by approved methods of present practice.

This does not detract from the investigation on autolysis, for which samples which proved to be not sterile (as noted) were rejected. It should be noted that we have observed that rather large amounts of fluid sometimes exude from meat *shortly* after irradiation. It seems, therefore, that other factors besides autolysis can cause exudation in irradiated meats.

Dr. RADOUCO-THOMAS (Switzerland): The fundamental issue is whether the muscle tissue of normal healthy animals is "sterile", and there are two conflicting schools of thought on this subject. From the microbiological point of view, however, the use of $\gamma$-radiation to treat meats would seem preferable.

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# Irradiation of Eggs and Egg Products*

J. BROOKS,† R. S. HANNAN‡ and BETTY C. HOBBS§

It has been known for some time that egg products may contain members of the *Salmonella* group capable of causing food poisoning, and the position has been rendered more serious by the recent discovery of *S. paratyphi* B in some of the samples of egg products imported into the United Kingdom. Attempts to destroy salmonellae in liquid egg by pasteurization before freezing or drying have met with considerable success but the process requires careful control. In any case, at the present time most of the frozen whole egg consumed in the United Kingdom has not been pasteurized. The tins in which the frozen product is distributed hold up to 20 kg, and are therefore of a size suitable for treatment with $\gamma$-radiation. After treatment of small samples of frozen whole egg with 2 MeV cathode rays it was concluded that a dose of about 0·3–0·5 Mrad would destroy the numbers of salmonellae normally encountered in the product without impairing the baking qualities of the material. Whole tins, each containing 10 kg of infected material, were therefore irradiated in the frozen state with $Co^{60}$ $\gamma$-rays. Two tins were treated at each of three dose levels of 0·3, 0·4 and 0·5 Mrad. No salmonellae were detected in duplicate samples of 25 g of material taken from each of the tins after irradiation. If the effectiveness of the treatment is confirmed, the process has obvious attractions since it dispenses with the need to thaw or otherwise to handle the product. The paper also contains a general discussion on the irradiation of eggs and egg products.

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## INTRODUCTION

Eggs in shell are not particularly susceptible to bacterial attack. Newly-laid eggs usually contain no bacteria and are armed with a defensive mechanism against casual infection from outside. Egg products, on the other hand, are usually kept and distributed in either the frozen or dried state, and under these conditions the bacteria that are present tend to die rather than to multiply. Nevertheless, bacteriological problems do exist. Shell eggs are not impregnable, and the production of dirty eggs and the washing of such eggs on the farm lead to the infection and rotting of some of them. Moreover, considerable care is required during the preparation of large volumes of liquid egg if an excessive multiplication of bacteria is to be avoided before freezing or drying. Furthermore, bacteria will start to multiply again if frozen or dried egg is thawed or reconstituted and kept at too high a temperature.

Much work has been done on these problems, and much is known about the conditions necessary to produce and market eggs and egg products with good keeping properties. Most of these conditions are not intrinsically difficult to fulfil although for a

* Most of the experimental details described in this paper have already been presented to the Second International Conference on the Peaceful Uses of Atomic Energy, Geneva, 1958.

† Low Temperature Station for Research in Biochemistry and Biophysics, Department of Scientific and Industrial Research, Cambridge.

‡ Low Temperature Station. Present address: The Metal Box Company Limited, Research Division, Acton, London.

§ Food Hygiene Laboratory, Central Public Health Laboratory, Colindale, London.

number of reasons they are not always fulfilled. A problem, however, which cannot be solved by the strictest care during manufacture is the presence of pathogens in a proportion of egg products. This can be seen from the fact that although a direct microscopic count will show whether or not the product has been exposed to grossly unhygienic conditions during manufacture, there is no relation between the magnitude of such counts and the presence or absence of pathogens. Consequently, most of the work on the irradiation of eggs has been devoted to the destruction in egg products of organisms capable of causing food poisoning, and chiefly of organisms of the *Salmonella* group.

As hen eggs, unlike duck eggs, had rarely been implicated in cases of food poisoning, and because organisms of the *Salmonella* group had rarely been encountered when individual hen eggs were examined,[1,2] considerable surprise was evoked when from 10 to 39 per cent of the samples of spray-dried whole egg prepared in North America and elsewhere during the second World War were found to contain salmonellae.[1,3,4] The reason for this widespread incidence appears to be: first, that the shells or the contents of a very small proportion of hen eggs are, in fact, infected with such organisms,[1,5,6] more particularly if the shells are dirty or have been washed or are cracked or leaking, and secondly, that as large volumes of liquid egg were accumulated during the manufacture of dried egg, there was ample opportunity for a few infected eggs to contaminate large volumes. Smaller volumes of liquid egg tend to be accumulated when frozen egg is manufactured. Furthermore, freezing tends to kill fewer bacteria than does spray-drying. Comparing the two processes, therefore, and with other things being equal, it might be expected that the proportion of samples of frozen egg contaminated with salmonellae would tend to be rather smaller but that the number of organisms in the positive samples would tend to be larger.

Attempts to destroy salmonellae in liquid egg by pasteurization before freezing or drying have met with considerable success,[7–10] and some North American, Australian and British plants and all Polish plants are now equipped with pasteurizers. The process, however, requires careful control,[9] and it has been reported[11,12] that, because of this, the process is not invariably successful under routine commercial conditions. Nevertheless, pasteurization greatly reduces the risk of infection. This fact, however, is of little more than academic interest at present to users of egg products in the United Kingdom since most of the products consumed, whether imported or home-produced, have not been pasteurized.

About 20,000 samples of egg products have now been examined by the Public Health Service[13] in this country. For frozen whole egg—an intimate mixture of the white and yolk and the principal product—and excluding samples which contained only *S. pullorum* (which is seldom pathogenic to man), it was found that 5, 10, 16 and 29 per cent respectively of Australian, Chinese, New Zealand and British samples contained salmonellae. The position was rendered more serious by the discovery[13,14] that 0·21 per cent of the samples of Chinese frozen whole egg (and 0·58 per cent of the samples of Chinese dried albumen) contained *S. paratyphi* B. Later examinations have shown a much higher level of contamination in some Chinese shipments. Furthermore, several outbreaks of paratyphoid fever in the United Kingdom have been proved by the Central Public Health Laboratory to have been caused by the consumption of cream cakes prepared in bakeries using Chinese frozen whole egg contaminated with *S. paratyphi* B of the same phage type as isolated from some patients and some members of the bakery staff.[14] In fact, the association of egg products with paratyphoid B fever has created a new situation in the epidemiology of the disease. These facts seemed to warrant a study of the effects of ionizing radiations with special reference to the feasibility of destroying salmonellae in large tins of frozen whole egg with $\gamma$-radiation.[15] Preliminary experiments were carried out with small samples of liquid or frozen whole egg and 2 MeV cathode rays.[15]

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## CATHODE RADIATION

The destruction of salmonellae in egg products by 3 MeV cathode radiation has been investigated by the Massachusetts Institute of Technology and collaborating laboratories. The first study[16] was concerned with liquid whole egg, and later ones with liquid, frozen and dried albumen[17] and with dried whole egg and dried and frozen yolk.[18] Small samples (2 g or 7 g) were irradiated after inoculation to the extent of several million organisms per gramme with one of three different species of salmonellae. With liquid whole egg, the average dose (recalculated as megarads) required to achieve a $10^7$-fold reduction in the number of salmonellae present was about 0·25 Mrad for *S. typhimurium* and *S. paratyphi*, and about 0·1 Mrad for *S. senftenberg*. A dose of 0·25 Mrad imparted an off-flavour and odour to the liquid which could be detected in scrambled eggs made from it but unflavoured sponge cakes made with it were comparable to control cakes in respect of volume, texture, flavour and odour.

The general conclusions drawn from the American work[16–18] were:

(a) salmonellae in dried egg products were more resistant to irradiation than the same organisms in the corresponding liquid or frozen product,

(b) a 40–50 per cent increase in dose would be expected to change the reduction in numbers from a value of $10^7$-fold to one of $10^{11}$-fold,

(c) only minor changes in flavour were apparent when unflavoured cakes were used to test the effect of a dose equivalent to a $10^7$-fold kill but highly significant changes were evident when scrambled eggs or custards were used for this purpose, and

(d) when an irradiated liquid was subsequently spray-dried, there was no significant difference between scrambled eggs prepared from the dried material and from control material.

It seems probable, therefore, that most of the compounds responsible for the off-flavours evaporate during baking or spray-drying.

TABLE 1. Presence or absence of salmonellae in frozen and thawed whole egg after irradiation with 2 MeV cathode rays

| Dose (Mrad) | Temp. of irradiation | | |
|---|---|---|---|
| | −70°C | −10°C | +15°C |
| 0·1 | +ve | +ve | +ve |
| 0·2 | +ve | +ve | −ve |
| 0·3 | +ve | +ve | −ve |
| 0·5 | −ve | −ve | |
| 0·75 | −ve | −ve | |
| 1·0 | −ve | −ve | |
| 1·5 | −ve | −ve | |

The results of our preliminary experiments with 2 MeV cathode radiation and liquid and frozen whole egg are summarized in Table 1. All the samples were taken from a tin of Chinese frozen egg naturally contaminated with *S. paratyphi* B, *S. thompson* and *S. pullorum*. Samples in thin layers in contact with air in sealed aluminium tins were irradiated whilst the temperature of the material was −70°C, −10°C and 15°C. The bacteriological examinations were made on duplicate samples of 25 g of material.[14] Three series of experiments were made with frozen whole egg, and two series with thawed whole egg; there was no difference between similar experiments at the same dose level.

The organoleptic changes resembled those reported by the American workers. The irradiated material had a slightly metallic odour and flavour, but this was not apparent in the finished sponge cakes. Nor was the baking quality, as judged by sponge cake volume, crumb texture and so on, in any way different from that of the unirradiated controls. Baking tests were made up to a dose level of 0·75 Mrad. The temperature of irradiation and the precise dose seemed to have little effect on the intensity of the irradiation odour of the raw material.

It will be seen that the borderline dose for the frozen whole egg lay between 0·3 and 0·5 Mrad, and for the thawed whole egg, between 0·1 and 0·2 Mrad. The latter range is rather lower than that reported by the

American workers for a $10^7$-fold kill in liquid whole egg inoculated with large numbers of *S. typhimurium* or *S. paratyphi* (about 0·25 Mrad). The difference may be partly due to the relation between the effective dose and the number of contaminating organisms. The number of salmonellae in the Chinese material (the MPN count was 21 organisms per g) was considerably smaller than in the American material, and the absence of salmonellae in duplicate samples of 25 g corresponds to an approximately $10^3$-fold reduction in numbers. At this point it is convenient to consider what are the numbers of salmonellae likely to be encountered in infected samples of commercial frozen whole egg. Unfortunately, the relevant data are very scanty but they suggest that the number may rarely exceed 1500 per g, and that the average number may be considerably lower, possibly of the order of 50 per g.

## GAMMA RADIATION

It is necessary, of course, to use $\gamma$-radiation for the treatment of large tins of frozen material. Such tins were irradiated in a refrigerated enclosure with $Co^{60}$ $\gamma$-rays. The temperature of the material was about $-10°C$. The results obtained with six different tins of contaminated Chinese frozen whole egg using doses ranging from 0·3 to 0·5 Mrad are set out in Table 2. Each tin contained 10 kg of material. The contaminants present before irradiation—three different species in all—are given in the first column of the table, and the dose in the second column. It will be seen that no salmonellae could be detected in any of the tins after irradiation.

Baking tests were made in the laboratory using a tin of uncontaminated Chinese frozen whole egg as control. Triangular tasting tests showed no significant preference for sponge cakes made with the control material or with material irradiated with a dose of 0·5 Mrad, although some tasters could distinguish between them. In a further experiment, four tins of frozen whole egg, each containing 7·3 kg of material, were prepared from a batch of English fresh eggs. Two were irradiated with $\gamma$-radiation, the other two served as controls. The dose was 0·5 Mrad. The baking quality of these materials was examined by three laboratories. All agreed that there was little or no appreciable difference between controls and treated material except that one of the laboratories detected a very slight foreign flavour in the sponge cakes prepared from one of the tins of irradiated material. This flavour was less apparent when the frozen egg was tested again after 1 week but remained at the same level after 2, 3 and 4 weeks storage in the frozen state.

TABLE 2. Presence or absence of salmonellae in tins of frozen whole egg after irradiation with $\gamma$-rays

| Original contamination | Dose (Mrad) | Salmonellae |
|---|---|---|
| *S. paratyphi* B | 0·3 | −ve |
| *S. newport* | 0·3 | −ve |
| *S. paratyphi* B | 0·4 | −ve |
| *S. newport* | 0·4 | −ve |
| *S. paratyphi* B | 0·5 | −ve |
| *S. thompson* | 0·5 | −ve |

When peroxide values of irradiated frozen whole egg were measured, there was evidence of a small peroxide after-effect.[19] As frozen egg is often kept for several months in the frozen state before use, it is possible that the flavour of the material might deteriorate slowly during such periods of storage. It is proposed, therefore, to test the material again after about 12 months storage.

A number of effects that were observed during the course of the work were briefly:

(a) irradiation reduced the number of micro-organisms other than salmonellae that were present,
(b) irradiation slightly bleached the colour of whole egg but the effect was much less than the natural variation of pigment concentration between different samples,
(c) irradiation greatly increased the weak fluorescence of liquid egg white in ultra-violet light, and
(d) the irradiation of frozen whole egg decreased the viscosity of the thawed material.

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## DISCUSSION

Naturally clean eggs seldom rot even when kept at ordinary temperatures for 3–4 weeks, and the proportion of rots after cold storage for 6 months is only about 0·5 per cent. Dirty eggs do not keep so well, and the washing of such eggs usually increases the incidence of rotting still further. The ill-effects of washing can be countered by heating the washed eggs in water or oil ("shell pasteurization" or "thermostabilization") although, as would be expected, the effectiveness of the treatment tends to decrease when the period between washing and treatment begins to exceed about 9 days. The surface of the washed shell could obviously be sterilized by irradiation but it would be necessary to increase the depth of penetration if the interval between washing and irradiation were unduly prolonged. It seems probable that penetrating radiations would also have to be used to make sure of destroying pathogens. Most of the salmonellae found within the occasional egg are probably derived from faecal contamination of the shell surface by an infected hen but infection of the liquid portion of the egg may also occur in the oviduct before the shell is formed. HANNAN[20] and others[21,22] have noted, however, that irradiation of the white imparts an off-flavour to it, and partly destroys the structure of the gel known as the thick white. For this reason, irradiation does not seem to be a promising method of improving the keeping qualities of shell eggs especially as other methods of doing so are available. An effective means is to dry-clean dirty eggs instead of washing them. Dry-cleaned eggs keep almost as well as naturally clean eggs.

As the proportion of hen eggs in shell that are infected with salmonellae is so small, and as most shell eggs, at least in this country, are usually cooked in one way or another, it seems unlikely that it would be worthwhile attempting to irradiate all hen eggs in order to obviate a health hazard which, as far as can be judged, seems to be very small. The egg products present a very different situation. The proportion of infected samples is by no means negligible. Moreover, these products are used by bakers, confectioners and caterers which necessarily widens the potential range over which cross-transfers of infection could conceivably occur. An effective means of destroying pathogens in egg products is therefore very desirable. The results described here and elsewhere by other workers suggest that this could be done by ionizing radiations.

Although attention has been paid in this paper to the feasibility of destroying salmonellae in large tins of infected frozen whole egg by irradiation, this is not the only possible way of dealing with such tins. It has been shown that infected frozen whole egg can be thawed and pasteurized with little damage to its baking properties.[23] Irradiation of the tins, however, has the advantage that it dispenses with the need to thaw or otherwise to handle the product. Nevertheless, either of these treatments might be regarded as something in the nature of a temporary expedient for in the long term it would clearly be more convenient to irradiate or pasteurize the liquid egg before it is frozen or dried. It is difficult at present, however, to assess the relative merits of the two processes. Leaving economics on one side, it is probably safe to say that irradiation has the advantage of providing a closer approach to a complete destruction of pathogens with less risk to the functional properties of the material whereas pasteurization has the advantage of producing no off-flavours or odours. Also to be taken into account when comparing the two processes is the fact that the organoleptic changes produced by irradiation have little or no effect on the taste and odour of baked goods prepared from irradiated egg products and the fact that, at least in this country, some frozen whole egg is used by caterers to prepare scrambled eggs and custards in which such irradiation odours and flavours can be detected.

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# The Present Status of the Irradiation of Foods in the U.S.A.—Eggs

B. S. SCHWEIGERT

American Meat Institute Foundation and Department of Biochemistry
The University of Chicago, Chicago, Illinois, U.S.A.

STUDIES at the University of California and at the Massachusetts Institute of Technology have indicated that flavor changes occur in scrambled eggs with dosages of 1·3 Mrads treatment. Spray-drying eggs after irradiation successfully eliminated any significant flavor differences attributable to irradiation. Relatively low dosages were required to destroy salmonella (0·125–0·6 Mrad). Other studies with a treatment of 0·6 Mrad showed no off-flavors in fried, scrambled, or hard-boiled eggs, but a stale flavor was observed in soft-boiled eggs. These eggs showed a change in form when broken from the shell. The albumin tends to thin and the whites tend to "run". No improvement in this undesirable effect was observed during storage.

It would appear that irradiation has little application for the preservation of fresh eggs, but may have some application in prepared products.

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# Some Effects of Ionizing Radiations on Liquid Whole Milk and Whey Protein

G. GLEW

U.K.A.E.A., Technological Irradiation Group, Isotope Division,
Wantage Radiation Laboratory, Wantage, Berks

Liquid whole milk was irradiated, and changes in flavour, alkaline phosphatase activity and acid soluble nitrogen were examined. Radiation induced flavour changes became detectable at 20,000 rads whereas 25 Mrads were required to inactivate the enzyme alkaline phosphatase. Preliminary experiments on the effects of combined heat and radiation treatment on phosphatase activity are described. Increased u.v. absorption of irradiated solutions of $\beta$-lactoglobulin and radiation induced changes in the electrophoretic behaviour of whey proteins were studied.

PROCTOR *et al.* have been studying the effects of radiation on milk for many years and have published a series of papers on the problems involved.[1–5] They have developed an irradiation procedure by which, it is claimed, sterile milk can be produced, indistinguishable in odour and flavour from fresh milk. Their method consists of irradiating milk *in vacuo* and removing off-flavour compounds as they are formed by simultaneous distillation. This work is of considerable theoretical interest but it is difficult to see how the method can compete with the well-established commercial pasteurization and sterilization processes. This paper will describe some of the results obtained by irradiating milk in sealed containers in an atmosphere of air.

## METHODS USED IN PRESENT TESTS

*Milk*

Samples of unpasteurized, bulked morning Jersey milk were obtained and treated within a few hours of milking.

*Irradiations*

Electron irradiations were performed in thin-walled 0·5 in. test-tubes revolving in the beam of a 4 MeV linear accelerator. Milk was also irradiated in standard McCartney bottles by $\gamma$-rays from a $Co^{60}$ source.

*Estimation of alkaline phosphatase activity*

Milk samples were incubated at 37°C for 15 min with disodium phenyl phosphate buffered at pH 9·7. Phenol liberated was estimated using FOLIN and CIOCALTEU'S reagent.[6]

*Electrophoretic pattern of changes in whey protein*

These measurements were made utilizing a horizontal electrophoresis chamber, barbiturate/acetate buffer (pH 8·6) and a voltage of 7 V/cm. Whatman No. 3 MM filter paper strips were used as the supporting medium and the protein was stained with 1% bromophenol blue in 90% ethanol saturated with $HgCl_2$.

*Estimation of acid-soluble nitrogen*

The protein was precipitated with 15% trichloracetic acid (wt/vol) and the tyrosine/tryptophan in the filtrate estimated using Folin and Ciocalteu's reagent. Samples were also digested at pH 2·0 for 15 min at 25°C with pepsin (final concentration 0·04%). The acid-soluble filtrates were again examined for tyrosine/tryptophan.

*Flavour tests*

These tests were conducted on samples of irradiated milk 1–3 hr after irradiation. A modified triangle test was used and fifteen tasters were selected, on the basis of consistency and discrimination of judgement, to take part in the trials.

## RESULTS AND DISCUSSION

*Inactivation of alkaline phosphatase by irradiation*

The enzyme inactivation curve is plotted semi-logarithmically in Fig. 1.

The curve is exponential. A dose of approximately 24 Mrads was necessary for complete inactivation. Electron dose rate, within the range 0·75–4·3 Mrads/min did not affect the magnitude of inactivation. The inactivation curve obtained using high dose rate $\gamma$-rays (produced by bombarding a platinum target with electrons) at a dose rate of 0·175 Mrad/min was not significantly different from the electron inactivation curve.

It is evident that the alkaline phosphatase of milk is extremely radio-resistant and that dose rate, within the range specified, has no effect on the rate of inactivation.

*The complementary effects of heat and radiation on alkaline phosphatase*

A number of workers in the microbiological field have found that radiation sensitizes some micro-organisms to heat treatment.[7,8] It has also been suggested that radiation sensitizes protein to de-naturation by heat.[9,10] Irradiating with $Co^{60}$ $\gamma$-rays at 40°C gives increased inactivation of the enzyme compared with irradiation at 20°C (Table 1) ($P = 0{\cdot}015$).

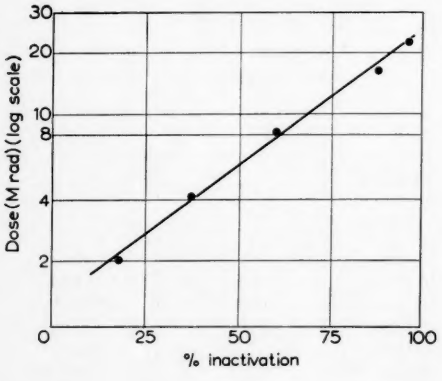


FIG. 1.

TABLE 1. The effect of temperature during $\gamma$-irradiation on phosphatase activity (dose rate: 0·18 Mrad/hr)

| Temp. (°C) | Dose (Mrads) | | | | | |
|---|---|---|---|---|---|---|
| | 0·25 | 0·5 | 1·0 | 1·5 | 2·5 | 4·6 |
| | % Inactivation of enzyme | | | | | |
| 20 | 5·0 | 8·5 | 17·9 | 23·4 | 30·4 | 53·8 |
| 40 | 2·2 | 9·1 | 23·2 | 30·1 | 44·3 | 61·6 |

Table 2 shows the effects of heating at 50°C for 15 min before and after irradiation. Irradiation before heating ($P = 0{\cdot}005$) is more effective in sensitizing the enzyme than heating before irradiation. This result is in agreement with results of work on micro-organisms in that a synergism exists only when irradiation precedes heating.

These results suggest that the inactivation of the enzyme can be significantly increased by the combination of heat and irradiation treatments described, although none of the treatments resulted in complete inactivation.

TABLE 2. The effect of heating at 50°C before and after irradiation on phosphatase activity
(Figures in "Heated" and "Unheated" columns represent % inactivation)

| Dose (Mrads) | Unheated | Heated | |
|---|---|---|---|
| | | Before irradiation | After irradiation |
| — | — | 4·9 | 6·8 |
| 0·25 | 2·1 | 5·9 | 4·7 |
| 0·50 | 5·0 | 10·1 | 13·2 |
| 1·00 | 13·1 | 18·6 | 22·0 |
| 2·00 | 23·6 | 21·9 | 31·3 |

*Irradiation-induced changes in the milk protein*

*Electrophoresis of whey protein.* Two series of experiments were performed.

(1) Whole milk was irradiated and the whey protein subsequently prepared for electrophoresis. Changes in α-lactalbumin became apparent at about 1·0 Mrad.

(2) Dialysed whey was prepared and irradiated prior to electrophoresis. Changes in α-lactalbumin commenced at about 0·5 Mrad.

In both experiments the α-lactalbumin band had disappeared at 2·0 Mrad. It was expected that the other milk components would protect the whey protein during irradiation under conditions described in experiment (1) and that the whey proteins would be more radio-sensitive when separated from the casein and fat as in experiment (2).

*Ultra-violet absorption spectra of irradiated β-lactoglobulin.* β-Lactoglobulin prepared by the method of ASCHAFFENBURG and DREWREY[11] was irradiated with a γ-ray dose of 3·81 Mrads at two concentrations, 17·3 and 32·3 mg/ml in 0·1 N NaOH. There was no shift of absorption maxima after irradiation and Table 3 gives the *E* values at the two absorption maxima.

Protein concentration affects the extent of change, greater change being observed at lower concentration. The compound (or compounds) causing the increased ultra-violet absorption is not removed after 20 hr dialysis against running distilled water. Increased ultra-violet absorption by proteins and amino acids has been described by many workers and MCLEAN[12] and CARROLL *et al.*[13] have suggested that a diphenyl-type linkage between several protein molecules may occur during irradiation. ALEXANDER *et al.*[14] have suggested that the apparent increase is due to loss of light by the greatly increased scattering by large aggregates formed during irradiation.

TABLE 3. Irradiation induced changes in absorption spectrum of β-lactoglobulin
(Protein dissolved in 0·1 N NaOH. Dose: 3·81 Mrads)

| Protein concentration (mg/ml) | Treatment | $E^{1\%}_{1\text{ cm}}$ 282 mμ | $E^{1\%}_{1\text{ cm}}$ 289 mμ | % increase at 289 mμ |
|---|---|---|---|---|
| 17·3 | control | 105·8 | 107·2 | |
| | irradiated | 181·8 | 170·1 | 58·7 |
| 32·3 | control | 109·0 | 111·3 | |
| | irradiated | 134·8 | 132·8 | 19·3 |

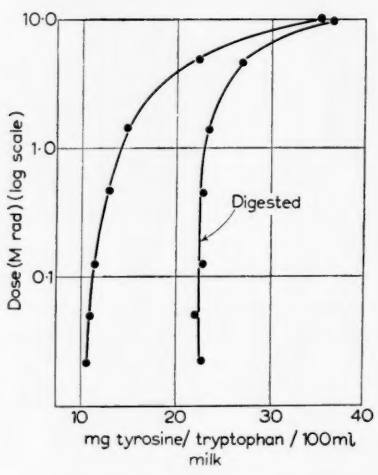


FIG. 2.

*Changes in acid-soluble nitrogen before and after peptic digestion.* Milk was irradiated with γ-rays at levels of 0·025–10·0 Mrads and the tyrosine/tryptophan in the acid-soluble filtrate estimated using Folin and Ciocalteu's reagent. Fig. 2 gives the results of this experiment. As the dose increases the protein becomes less digestible by pepsin.

*Flavour studies.* Milk treated with doses of 15, 20, 25, 30, 40 and 50 krad/γ-rays was presented in the form of a modified triangle test. Tasters were asked to score differences

TABLE 4. Some results of flavour tests on irradiated milk

| Dose (rads) | *t* | Probability |
|---|---|---|
| 50,000 | 3·854 | < 0·001 |
| 40,000 | 3·074 | < 0·01 |
| 30,000 | 2·950 | < 0·01 |
| 25,000 | 2·047 | < 0·05 |
| 20,000 | 2·985 | < 0·01 |
| 15,000 | 0·264 | 0·8–0·9 |

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between samples on a numerical scale from 1 to 10. Table 4 gives the results of these experiments.

At all treatment levels above 15 krads significant differences between the flavours of untreated and irradiated milk were observed. It appears that the dose at which irradiation flavour becomes detectable is between 15 and 20 krads.

*Conclusions*

The irradiation of milk, because of its complex chemical composition, poses many problems. The present studies are being undertaken not so much in an attempt to achieve a commercial process, but because data on the changes that take place should yield information which may be of use in application to other foodstuffs. The results presented here do not, as yet, indicate any clear applications for the use of radiation in preserving milk.

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# Bacteriological Investigation of Liquid Milk Irradiated in Gamma-source

BARBARA M. FREEMAN

Technological Irradiation Group, Isotope Division, Wantage Radiation Laboratory, Wantage, Berks

(1) The effect of irradiation of liquid milk on the bacterial flora is discussed.
(2) Marked reductions in the total counts and coliform counts is shown at varying dose levels.
(3) Spore-formers surviving irradiation treatment increased during the storage period and appeared between the fourth and eleventh days after irradiation.
(4) The occasional odd colonies appearing after the higher irradiation doses could not be classified as truly resistant organisms.

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## INTRODUCTION

UNPASTEURIZED, morning Jersey milk was used in this investigation, to find the dose range necessary to obtain commercial sterility. The samples were obtained as soon as possible after milking and were irradiated in a $Co^{60}$ source at 20°C.

Bacteriological investigations were carried out within a few minutes of irradiation.

## METHODS

The milk was received in pint bottles and mixed thoroughly, by shaking in a sterile flask, before pouring into McCartney bottles for irradiation. The dose levels chosen for the initial series were 0·25 Mrad; 0·5 Mrad and 0·75 Mrad.

Nine samples of milk were irradiated at each dose level—the number of samples being limited by the amount of space available in the radiation source. Nine aliquots, to be used as unirradiated control samples, were poured at the same time. These controls were placed near the irradiation cell during irradiation of the other samples, in an attempt to keep all samples at as near an equal temperature as was possible under existing conditions.

One sample from each dose level, with a control sample, was tested on the day of irradiation and on each subsequent day, omitting the week-end. During storage, all samples were kept in water, in the 22°C incubator.

Total counts were done on milk agar, coliform counts on Difco MacConkey agar, coliform tests in MacConkey broth, and 1 ml of each sample inoculated into litmus milk. All tests were incubated at 37°C for 48 hr.

Each sample was also plated on Wort agar, incubated at 22°C for 3 days and examined for the presence of moulds and yeasts.

All tests were carried out immediately after irradiation on the first day. On the following days, the tests were done at 2.0 p.m. in the first week and 11 a.m. in the second week.

A further set of experiments was carried out in the same way, but with increased dose levels, 0·75 Mrad; 1·0 Mrad and 1·25 Mrad.

## RESULTS

The results discussed here are from tests carried out over a period of 12 weeks.

During this time, the initial total counts on the unirradiated control samples were between $1{\cdot}8 \times 10^3$ orgs/ml and $7{\cdot}3 \times 10^5$ orgs/ml, with coliform counts varying from 25 coli/ml to $6 \times 10^4$ coli/ml, and positive coliforms in $10^{-2}$ to $10^{-5}$ dilutions. Each control sample produced a typical acid clot in litmus milk.

The initial count of moulds and yeasts was very low throughout the twelve-week period, from 0 to 12/ml.

Throughout the testing period, the survival of organisms after irradiation was <0·1 per cent of those originally present. With the lowest irradiation dose (0·25 Mrad) a few organisms survived the treatment, but always less than 10 orgs/ml. At the higher dose levels, occasional odd colonies appeared, but never more than 1 org/ml.

The total counts on milk agar increased gradually over the period of testing, in the samples irradiated at the lowest dose level. By the eighth day after irradiation, these counts ranged from 4 orgs/ml to $1 \times 10^5$ orgs/ml and were mainly organisms of the spore-bearing type.

TABLE 1. Total counts, 1st day

| | Unirradiated control | $0{\cdot}25 \times 10^6$ rads | $0{\cdot}5 \times 10^6$ rads | $0{\cdot}75 \times 10^6$ rads |
|---|---|---|---|---|
| 1 | $1{\cdot}1 \times 10^4$ | 4 | <1 | 0 |
| 2 | $2{\cdot}0 \times 10^4$ | 5 | 0 | <1 |
| 3 | $>3{\cdot}0 \times 10^4$ | 0 | <1 | <1 |
| 4 | $3{\cdot}5 \times 10^3$ | 2 | 0 | 0 |
| 5 | $7{\cdot}0 \times 10^3$ | <1 | 0 | 0 |
| 6 | $1{\cdot}1 \times 10^4$ | 1 | 1 | 0 |
| 7 | $6{\cdot}6 \times 10^3$ | 7 | 0 | 0 |
| 8 | $6{\cdot}5 \times 10^3$ | 10 | <1 | 0 |

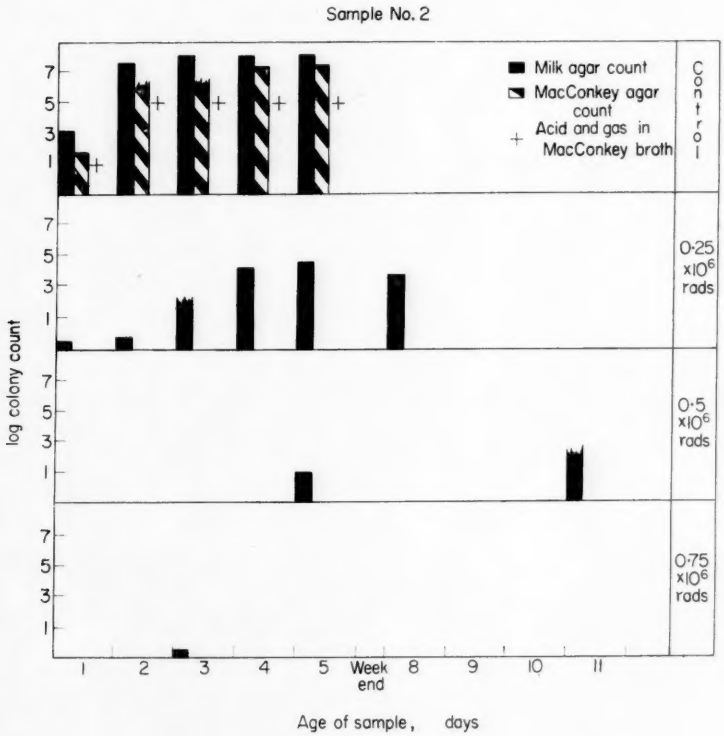


FIG. 1.

At the higher dose levels, a few spore-bearing organisms appeared sporadically and developed after several days storage at 22°C. At 1 Mrad and 1·25 Mrad dose levels, the counts never increased after storage, but always remained below 1 org/ml. Whether any organisms survived the higher dose levels of irradiation will have depended upon the numbers and types present originally.

Counts on MacConkey agar and coliform tests were negative at all dose levels immediately after irradiation. With the lowest dose level, a low count occasionally appeared after a few days storage. No positive coliforms were found at the higher dose levels ($\geqslant$0·75 Mrad) throughout the testing period (Figs. 1 and 2 are examples of results from two separate milks.)

None of the irradiated samples produced acid or clot in the litmus milk at any time during testing. At the lower dose levels (0·25 and 0·5 Mrad) sometimes the milk was alkaline and digested, due to survival of Gram-positive bacilli.

No moulds or yeasts were shown at any dose level immediately after irradiation. With the samples irradiated at 0·25 Mrad the count of moulds and yeasts increased suddenly from the third to fifth day of storage. It appears that this dose level is insufficient to kill the moulds and yeasts, yet high enough to kill the lactobacilli and coliforms, producing conditions in the milk suitable for the development of any moulds or yeasts which may have been present in the original raw milk and survived the irradiation.

An attempt was made to type some of the organisms surviving irradiation; the majority of these were Gram-positive spore-bearing bacilli, including *B. subtilis*, *B. licheniformis* and *B. megaterium* and a few micrococci. A number of the organisms

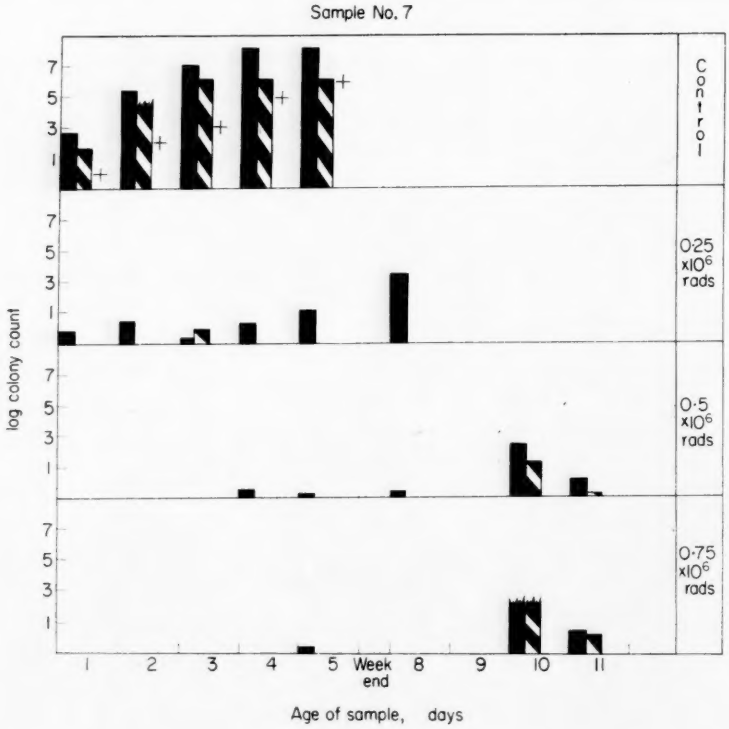


FIG. 2.

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surviving the higher levels of irradiation (1·0 and 1·25 Mrad) were irradiated again, as pure cultures in 10 ml sterile milk. None of these organisms survived the second irradiation and could not be classified as truly radiation-resistant.

## DISCUSSION

The irradiation dose levels in the initial experiments were not high enough to produce a completely sterile product, although apparent sterility was attained immediately after irradiation. When the dose levels were increased to 1·0 Mrad and 1·25 Mrad the results, over a period of eleven days storage at 22°C, showed that "commercial" sterility of raw milk may be reached at this level of irradiation.

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# The Present Status of Irradiation of Foods in the U.S.A.—Milk and Milk Products

B. S. SCHWEIGERT

American Meat Institute Foundation and Department of Biochemistry
The University of Chicago, Chicago, Illinois, U.S.A.

MILK is perhaps the most sensitive of all foods to flavor and odor changes that occur during irradiation. Considerable work has been done on the stability of the milk proteins, as well as changes that occur during irradiation of whole milk, skim-milk, cheese products, etc. The flavor changes can best be described as being similar to those that occur on prolonged heat treatment. Dosages of 7000–25,000 rads are sufficient to permit detection of off-flavors in milk or cream. Prepasteurization and homogenization of milk tends to increase the resistance to flavor changes. Somewhat higher dosages are required for flavor changes to occur if dried skim or whole milk is irradiated before reconstitution. If irradiated after reconstitution, the results are similar to those for fresh milk.

The group at Massachusetts Institute of Technology considers that the TBA test (thiobarbituric acid) is a good index of radio sensitivity for milk. While this test probably does not measure the off-flavor components, it appears to correlate with organoleptic tests. The components formed that are associated with undesirable flavors appear to arise from both the milk fat and from the fat-free residue. These results appear to be somewhat different from those for meat in which studies to date indicate that off-flavor constituents arise primarily from the lean of meat and not from the fat.

Considerable destruction of vitamins A and C and of tocopherols has been observed when milk is irradiated. Approximately 70–80 per cent of these vitamins are lost with irradiation dosages of 0·6–0·8 Mrad, while the activity of the enzyme phosphatase is reduced only 10 per cent.

A unique approach for eliminating the off-flavor constituents from milk during irradiation has recently been developed by PROCTOR *et al.* at Massachusetts Institute of Technology. By irradiating milk at low temperatures with concurrent distillation to remove volatile off-flavor constituents, it has been possible to produce a sterilized product by irradiation, with a high acceptability. This experimental finding is most significant and suggests another way that off-flavor and odor problems may be circumvented.

## CONCLUDING COMMENT

The varying results obtained on the acceptability of different food products given the same radiation dosage, when tested either after irradiation or storage, merit some comment. The training and experience of the taste panels are important factors, as well as variations in the pretreatment of the foods prior to irradiation that are difficult to control or to evaluate.

The food habits and background of the taste testers are undoubtedly important. Perhaps of greatest importance is realizing whether the panel is a trained analytical panel or is set up as a consumer-type panel. The latter, of course, is not expected to pick up small differences as readily as the trained panel.

As our knowledge increases we should be



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able to understand these factors to a greater degree and thus delineate the most fruitful products to consider for civilian use. In addition, we should be able to "pin-point" the long-range research effort through understanding the dosage rates required for sterilization under different circumstances, the nature of the chemical changes associated with off-flavors and odors, and techniques to circumvent the development of these off-flavors.

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# Discussion

Dr. KRAYBILL (U.S.A.): Have you examined the casein of milk by paper electrophoresis? What observations did you make of the colloidal stability of irradiated milk samples?

Mr. GLEW (U.K.): We have had no success with paper electrophoresis of casein. We have observed that samples irradiated with doses greater than 5 Mrads did gel after storage of upwards of 4 weeks at 30°C. We have not observed gelling in samples given lower doses of irradiation or stored at lower temperatures.

Dr. MOCQUOT (France): Have the effects of ionizing radiations on the enzyme xanthine oxidase in milk been examined, and have any measurements of the rennet coagulation time been made?

Mr. GLEW (U.K.): I have not examined the inactivation of this enzyme, but American work has shown that the dose at which complete inactivation is achieved is similar to that of milk alkaline phosphatase. The rennet coagulation time for milk has not been examined.

Dr. KON (U.K.): It would appear that there is little hope of practical application of ionizing radiations to the commercial treatment of milk. The fundamental aspects of this procedure are of great interest and in this connexion I would like to ask two questions:

(1) We have heard something about the effects on the nutritive value of milk and perhaps the specialists present might enlighten me further on this point.

(2) It is well known that exposure to visible light in the presence of atmospheric oxygen initiates a chain of rapid chemical changes in milk involving riboflavin, ascorbic acid, fat and the organoleptic properties, and I would like to know the effect of ionizing radiations when applied to deaerated milk that has not been exposed to light.

Dr. SCHWEIGERT (U.S.A.): The loss in vitamin content in milk is similar to that observed for other foods that have received similar irradiation treatment. The major losses are due to oxidative changes and include breakdown of tocopherols and vitamin A, ascorbic acid, thiamine and riboflavin.

Dr. STOSIĆ (Yugoslavia): Experiments have been carried out in Yugoslavia to determine whether ionizing radiations can be used to reduce microbial spoilage in certain kinds of cheese. It was found that complete sterilization could be obtained by doses of $1{\cdot}5 \times 10^6$ rads while at the dose of $0{\cdot}2 \times 10^6$ rads about 90 per cent of the organisms were inactivated. However, at doses as low as 25,000 rads adverse changes in the odour and taste (described as metallic in nature) occur in the cheese.

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# The Irradiation of Ware Potatoes*

W. G. BURTON† and W. H. de JONG‡

$\gamma$-Irradiation at 10,000 rads or even less will prevent the sprouting of potatoes, and its use on the commercial scale appears to be technically feasible. Its widespread employment on individual farms would perhaps be difficult and it has other disadvantages. For instance it may increase fungal rotting, and it accelerates the sweetening and tissue breakdown which normally occur after prolonged storage.

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We can only assess the present status of the irradiation of potatoes by defining precisely what we wish to achieve by irradiation and then by determining what we have in fact achieved. In broad terms of course, what we are endeavouring to do is to extend the storage life of the potatoes, but it is of no use to extend the storage life if by doing so we reduce the quality, or spend more than the saving justifies. For example, there is no doubt that the storage life of potatoes may be much prolonged by the use of refrigeration to hold them at a temperature which, depending upon the variety and other circumstances, may be from 2° to 5°C. In many cases, however, the cost of such storage might be regarded as uneconomic; and in any case the quality is so impaired by sweetening that refrigeration must be regarded as undesirable.

We may group under six main headings most of the relevant considerations to be borne in mind when discussing the status of the irradiation of potatoes.

(1) What desirable properties do we wish to be sure of retaining in the stored potatoes?
(2) What is the magnitude of the loss of potatoes during storage?
(3) Does irradiation reduce this loss and at what dosage?
(4) Are there any undesirable side effects?
(5) What is the net result of irradiation, taking all its effects into account?
(6) If the use of irradiation is desirable, is it practicable?

1. *The qualities which are considered desirable*

Consumers in different countries differ in their assessment of the various attributes of the potato which make up its quality—particularly in their reaction to the texture of the cooked potato and to the whiteness or yellowness of its flesh. There is general agreement however that black discoloration in the flesh is undesirable, as are a soggy texture, flavour taints, loss of nutrients, and—very important in the case of fried chips and crisps—a sugar content such that the product is too brown.

2. *The loss of potatoes during storage*

This is due to three main causes: disease, water loss, and sprout growth. The first of these is probably in general the most important, although it is difficult to give any estimate of overall losses, which vary greatly from year to year. As an example we may quote estimated storage losses of up to 50 per cent from late blight (*Phytophthora infestans*) in East Anglia in the storage season 1941–2, while in 1942–3 the losses from this cause averaged only some 3 per cent.[1] Irradiation is not regarded as a means of reducing losses caused by disease and under this heading we will not discuss them further. Water loss and sprout growth, although they vary somewhat from year to year, are much more predictable than disease losses. On the average it has been estimated that sprout growth on late stored potatoes in this country may range from about 0·5 per cent

* Ware potatoes are those sold for human consumption.

† Ditton Laboratory, Larkfield, Kent.
‡ Institute for Storage and Processing of Agricultural Products, Wageningen, Netherlands.

by weight in March to about 7 per cent in July. Water loss has been estimated to average about 0·5 per cent per month until February and then to increase up to some 2–3 per cent per month in May and subsequent months.[2] The loss attributable to sprout growth on ware potatoes alone in Great Britain must be something of the order of 30,000 tons, and water loss perhaps about 35,000 tons, of which 5000 tons are attributable to sprout growth because tubers lose water more rapidly when they have sprouted. The complete suppression of sprouting would therefore save, on the average, about 35,000 tons of ware potatoes a year in Great Britain.

3. *Reduction of the loss by irradiation*

Many workers have shown that sprout growth can be completely suppressed by irradiation with $\gamma$-rays[3], electrons[4] or X-rays.[5] The minimum dose to achieve suppression has not been precisely defined and actually there is evidence that this minimum dose may vary with the variety of potato,[6] with the physiological state of the tuber,[6] and also with the subsequent storage conditions. The amount of absorbed radiation necessary to prevent sprouting also differs with the different types of irradiation. For instance work by W. F. van Vliet of the Institute for Storage and Processing of Agricultural Products at Wageningen in conjunction with Philips, Eindhoven, has shown no depression of sprouting by irradiating at 10,000 rads with a 500 kV electron source, while a similar amount of energy (11,000 rads) in the form of X-rays from a 200 kV source caused complete sprout inhibition. $\gamma$-Irradiation will stop sprouting at a dose of the order of 10,000 rads and complete suppression has in fact been achieved in one case at a dose as low as 3000 rads using a 100-c $Co^{60}$ source.[7]

4. *Side effects of irradiation*

It is to be expected that a treatment which prevents sprouting may also have other, possibly undesirable, effects. The undesirable effects which have been reported as being caused by the comparatively low doses of $\gamma$-radiation necessary to prevent sprouting are loss of ascorbic acid, sweetening, increased susceptibility to storage rots, and final breakdown of the tubers.

(a) *Loss of ascorbic acid.* Irradiation with 10,000 rads causes a practically immediate loss of ascorbic acid—a 20 per cent loss has been observed after 2–3 hr.[6] During subsequent storage the rate of loss from irradiated tubers is at first more rapid than that from non-irradiated tubers, but tails off more rapidly. By the end of June there may be little difference between the ascorbic acid content of irradiated and non-irradiated tubers.[6]

(b) *Sweetening.* Irradiation increases the sugar content of the potato. The increase is greater the greater the dose but is considerable even at a dose of 3000 rads. In experiments at the Ditton Laboratory the sugar content has reached a maximum of about 1·5 per cent of the fresh weight some five days after irradiation with 10,000 rads. Thereafter there is a rapid fall in sugar content, and in the case of potatoes irradiated immediately after harvest there will be little difference in this respect between irradiated and non-irradiated tubers after storage for about two months. After prolonged storage non-irradiated potatoes sweeten very considerably. This sweetening appears to be accelerated by irradiation and in some cases a sugar content reached by non-irradiated potatoes in 10 months may be reached 6 months after irradiation, and there have been cases where irradiated potatoes have been inedible 10 months or so after harvest and irradiation.

(c) *Increased susceptibility to storage rots.* There is evidence that irradiation with 10,000 rads may increase the susceptibility of potatoes to infection by, for example, *Phytophthora* and *Fusarium* because of the prevention of wound periderm formation.[6] The increased susceptibility is temporary (less than 12 days), as despite the lack of periderm tissue the cells at the wounded surface become suberised.[6] Irradiation with 5000 rads apparently does not increase the susceptibility to fungal invasion.

(d) *Final breakdown.* In experiments at the Ditton Laboratory on the storage of irradiated

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potatoes at a temperature of 10°C, storage life has been terminated after some fifteen months by complete breakdown of the tubers.[7] The sweetening mentioned above may be an early symptom of this breakdown.

5. *The net result of irradiation*

The effect of irradiation is thus to stop sprouting completely but not to prolong storage indefinitely. There may be some increase in storage rots; there may be sufficient sweetening after six months or more to render the potatoes unacceptable, first for frying as chips or crisps, and later for other culinary purposes; and after fifteen months or more there may be complete breakdown of the tuber. The loss of ascorbic acid may not be serious, as late-stored non-irradiated potatoes would have but little more ascorbic acid than would late stored irradiated potatoes. Irradiation at a low level—say 5000 rads—either does not induce harmful side-effects, or does not induce them so soon, but it may not always stop sprouting completely. There seems little doubt however that irradiation at such a low level could well give commercially acceptable results until late spring or early summer—by which it is meant that the sprouting would be so slight as not to represent an appreciable wastage or impede the grading of the potatoes for sale.

6. *Whether large-scale irradiation is practicable*

We have pointed out what seem to be the limitations on the use of irradiation as a means of preventing sprout growth. Quite apart from these, before a method is introduced into agricultural practice we must examine the question as to whether it is practicable. As an example we will take conditions in Great Britain.

The tonnage of potatoes necessarily stored sufficiently late to benefit from a means of sprout suppression is rather more than one million tons. This tonnage is harvested when weather conditions are suitable over a period of about two months. There are about 80,000 commercial potato growers in Great Britain and any of these may store a proportion of his potatoes late if he thinks it to his financial advantage to do so. The number who actually store any quantity late is much less than this, perhaps about 5–10,000. If we wished to irradiate the potatoes which were to be stored late by these 5000 or 10,000 growers we should have either to provide transport to various irradiation centres or we should have to provide irradiation facilities at 5000 or 10,000 widely separated places, where potatoes were being lifted often at no very great rate—perhaps from 10 to 30 tons per day. The provision of irradiation facilities in such a system would seem to present many more problems than in the Netherlands where the facilities might be installed in the big co-operative potato stores.

Assuming that irradiation is feasible in the agricultural system as it exists, or that the system can be changed to make irradiation feasible, it remains to decide on the method of irradiating. In this connexion it is of interest that a prototype commercial irradiator is being tested in the U.S.S.R.[6] This consists of a concrete bunker beside one wall of the potato store. In the bunker are pipes which can be charged, with say, $Co^{60}$ from a mobile container, and the potatoes are fed by conveyor belts from the store into the bunker and back again. Some such method would seem to offer a reasonable promise of being commercially feasible.

We may conclude by summing up the present status of the irradiation of ware potatoes somewhat as follows:

There seems to be no doubt that the irradiation of potatoes to prevent their sprouting is technically feasible, though it may be that co-operative storage is better adapted to the employment of irradiation than is the practice of storing potatoes on individual farms. The irradiation dose should be the minimum which will give commercially acceptable sprout suppression —certainly less than 10,000 rads. Having said this it must be realized that irradiation is not a panacea for all storage ills. It does not reduce, and may even increase, fungal rotting. Although on the one hand it delays the end of the storage life by preventing

sprouting, on the other hand it hastens that end by accelerating the final sweetening and breakdown. The desirability of using irradiation to suppress sprouting must depend on the balance struck between these opposing effects.

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# Irradiation of Root Crops

KNUT MIKAELSEN

The Agricultural College of Norway, Institute of Genetics, Vollebekk

Moderate doses of $\gamma$-radiation (5000–15,000 rads) seem to be effective in preventing or inhibiting sprouting, growth and weight loss in various root crops, but the incidence of rotting seems to be a serious problem in irradiated material.

The experimental results seem to indicate that radiation treatment increases the susceptibility to rot infections. This problem seems to be solved in carrots, where minimum rot development was obtained when washed and clean roots were irradiated and stored in plastic bags and kept at cool storage temperatures.

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THE problems in irradiation of root crops are very similar to those involved in potato irradiation. It seems quite obvious that sterilization doses of several megarads are of no advantage and can hardly justify practical application. Doses of 2 Mrads in carrots influence the flavour and cause some bleaching and softening.[1,3,4]

Moderate or low doses of radiation for prevention or inhibition of sprouting seem to be successful since by inhibition of sprouting, storage life may be considerably extended. Although much work along these lines is in progress in many countries, there seems to be little published information on sprout inhibition for products other than potatoes.

Since there is little literature on the subject, the only material which has been available to us comes from the results of our own experiments carried out at the Agricultural College of Norway.

*Carrots*

The first experiment was started in 1954 with carrots. The roots were irradiated with 5000, 10,000 and 15,000 rads of $\gamma$-radiation and stored at a room temperature of 12–15°C. This high temperature was used to promote maximum growth and rotting. The carrots were stored in moist sand in wooden boxes.

TABLE 1. Effects of $\gamma$-rays on growth and rotting in carrots during storage

| Dose (rads) | Average length in cm of leaves at months from harvest | | | Per cent rotting at months from harvest | | |
|---|---|---|---|---|---|---|
| | 4 | 6 | 8 | 4 | 6 | 8 |
| 0 | 1·5 | 2·5 | 3·6 | 8 | 21 | 47 |
| 5000 | 0·6 | 0·8 | 1·0 | 7 | 20 | 47 |
| 10,000 | 0 | 0 | 0 | 10 | 25 | 58 |
| 15,000 | 0 | 0 | 0 | 12 | 35 | 65 |

During storage, measurements on growth or sprouting and observations on rotting were carried out 4, 6 and 8 months after harvest. No growth was observed in the carrots exposed to 10,000 and 15,000 rads, as late as 8 months after storage. At a dose of 5000 rads, some sprouting was noticed, but a marked and significant growth inhibition, as compared with the untreated controls, was observed. After 8 months new root hairs appeared in the untreated controls, whereas in the irradiated carrots root hair growth was completely absent.

Observations on rotting seem to indicate that a radiation dose of 5000 rads had no influence on rotting throughout the experimental period. At doses of 10,000 and 15,000 rads increased numbers of rotten

carrots were observed after 8 months of storage and it appeared that the infections came from an initial rotting in the dead tissue at the growing point of the carrots.

Further experiments were carried out in the years 1955/57 to test the significance of these results and to find out if other storage procedures or conditions could decrease rotting of irradiated carrots during storage.

In these experiments, the carrots were washed and cleaned after harvest, and packed in plastic bags which should protect the roots from infections from the outside. The carrots were then exposed to 5000, 10,000 and 15,000 rads of $\gamma$-radiation. After the radiation exposures the carrots were held in cool storage at 2–5°C throughout the experimental period. Analyses of growth, weight loss and rotting were carried out and the results after 6 months of storage (Nov.–May) are listed in Table 2.

TABLE 2. The effect of $\gamma$-radiation on growth, weight loss and rotting of carrots stored for 6 months in plastic bags at 2–5°C

| Treatment | Relative growth | Per cent weight loss | Per cent rotting |
|---|---|---|---|
| Untreated control | 100 | 38 | 53 |
| 5000 rads | 25 | 36 | 21 |
| 10,000 rads | 14 | 33 | 9 |
| 15,000 rads | 0 | 56 | 73 |

It appears from Table 2 that leaf and root hair growth are considerably reduced and inhibited in the roots exposed to 5000–15,000 rads of $\gamma$-radiation while at a dose of 15,000 rads, growth is completely prevented. Interesting results were obtained on the relation between radiation dosage, extent of rotting and of loss of weight during subsequent storage of the carrots. At dosages of 5000 rads and especially at 10,000 rads, the incidence of rotting was considerably reduced and, in the case of carrots treated at 10,000 rads, there was a slight but significant reduction in the loss of weight. At dosages of 15,000 rads, there was a pronounced increase in rotting which was probably responsible for the increased loss of weight from carrots treated at this dosage.

The high incidence of rotting in the roots where growth is entirely prevented is interesting. Probably damage to or the death of the outer cell layers had reduced the resistance of the carrots to rot development.

From the data given in Table 2 it would seem that a moderate dose of 10,000 rads is preferable since at this dose the minimum weight loss and rot infections were observed. At this moderate dose, the tissue is not killed, but is so seriously damaged that growth is considerably inhibited, though it was still sufficient to prevent rot infection from developing.

By comparing the results from the two types of experiments presented in Tables 1 and 2, it is obvious that sprouting can be easily inhibited or completely prevented at storage temperatures from 2°C to 18°C. Significant reduction in weight loss may also be obtained especially when the roots are stored at low temperatures.

Reduced resistance against rot infections seems to be a serious problem in irradiated carrots, especially when the roots are stored at high temperature (Table 1). However, from the experimental results listed in Table 2, it seems possible to prevent rot development when the roots are packed in plastic bags and stored at low temperatures.

*Other root crops*

Storage experiments have also been carried out with red beets, various turnips, Jerusalem artichoke (Topinamber) and onions.

With moderate doses of 5000–15,000 rads of $\gamma$-radiation, inhibition and prevention of sprouting have been obtained in all these crops. The radiation treatment extended storage life, but incidence of rotting was high. Similar experience has been reported by DALLYN *et al.*[2] with onions. Since the incidence of rotting was fairly high in the control samples, it may be of great importance to use almost non-infected material for radiation treatments, because sterilizing doses are not applied.

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# Potatoes and Root Crops

G. TRIPP*

Quartermaster Food and Container Institute for the Armed Forces, Chicago, Ill., U.S.A.

THE prevention of potato sprouting by radiation has received considerable attention in the U.S. Since the previous speakers have thoroughly covered the subject my remarks will be brief.

One of the most important factors in the irradiation of potatoes has been the interference with periderm formation. This interference of wound healing has in part been responsible for high losses in some lots of irradiated potatoes. We have found that it is desirable to store potatoes for a short period prior to irradiation to permit healing of wounds inflicted during harvesting.

During the past few years we have irradiated large quantities of potatoes for animal feeding studies. Sizeable quantities of sprout inhibited potatoes of the 1956 crop are still in good condition. These are Russet Burbank tubers which were treated with relatively low doses of radiation in the range of 7500–10,000 rads. Several tons of potatoes from the 1957 crop are in excellent condition. In both these lots sprouting, shrinkage and decay are negligible.

At this point it is well to consider the practical side of potato irradiation. The process is workable and produces satisfactory potatoes. However, this is not enough. The process must also either do something that cannot be done by other means, or do something better than another process, or perform the function at a lower cost. If the irradiation process, in addition to producing the desired result, does not meet one of the preceding conditions, it will not have any particular advantage over present methods. In the case of sprout inhibition, a chemical method exists which is satisfactory. The ultimate process will be determined by the results achieved by each method and the cost. The purpose of the United States Army Radiation Center is to secure cost data on a pilot plant scale. The operation of the USARC will aid materially in determining the feasibility of the process. It should be noted that these considerations apply to all foods processed by radiation.

In respect to other root crops, such as onions, some work is in progress. However, the preliminary nature of the results do not warrant comment.

* FAO Consultant.

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# Possible Applications of Ionizing Radiations in the Fruit, Vegetable and Related Industries

I. D. CLARKE

Technological Irradiation Group, Isotope Division, Wantage Radiation Laboratory, Wantage, Berks

Some effects of $\gamma$-radiation have been observed on the texture, flavour and processing properties of fruits and vegetables.

An appreciable reduction in mould decay of strawberries during storage at 20°C and 1°C has been observed after treatment with 0·2 to 0·4 Mrad. Flavour was affected slightly and the viscosity of the juice was reduced considerably.

The viscosity of juice extracted from irradiated whole berries, was reduced considerably by doses between 0·4 and 0·6 Mrad without critically affecting the flavour of the juice. The irradiated berries were free from a significant number of mould spores. No immediate commercial application was foreseen in the blackcurrant juice industry.

Plums were shown to be free from mould after a treatment of 0·4 Mrad and the initial spore count of apples is considerably reduced by a dose of 0·2 Mrad.

Some evidence has been obtained that the softening of pears can be delayed by a dose of 0·3 and 0·5 Mrad.

The published literature on the effects of radiation on other fruits and vegetables has been reviewed and commented upon.

## INTRODUCTION

Decay of fruits and vegetables is caused by two primary agencies:

(1) Microbial attack

(2) Chemical changes.

Microbiological attack can be classified further into bacteriological and mycological problems. In the case of the fruit industry, there is no bacteriological problem for the pH of the fruit juice is sufficiently low to inhibit the growth of bacteria. Needless to say, moulds and yeasts can develop rapidly on the substrates provided in fruit extracts. In the case of vegetables, bacteria and fungi can cause difficulties in storage.

Examples of decay caused by chemical changes, which in living tissues are catalysed by enzymes, can be readily found in the storage of fresh fruit and vegetables e.g. internal breakdown of pears, physiological diseases to which apples and pears are prone, the gradual development of a bitter flavour in stored carrots.[1,2,3]

The importance of (1) and (2), with regard to the storage life of any one fruit in any one year may vary quite considerably. As an example, the situation which arises occasionally with South African peaches may be quoted, where fungal attack is the usual cause of losses but occasionally a ship load of peaches will be unloaded at the docks, sold to the retailer and after about four days at ambient temperatures will be useless for sale due to the internal, physiological breakdown characteristic of low temperature injury.[4]

Physical changes, such as wilting and dehydration, may also determine the storage life of fruits and vegetables, but these can normally be reasonably minimized by adequate control of the environment.

Obviously, the storage life of plant materials is dependent upon many interacting factors, only a few of which have been enumerated in the previous paragraphs, and before the value of ionizing radiations can be fully realized, the inter-relationships between radiation treatment and all these other factors will have to be determined accurately.

## RESULTS

*Some Observations on the Effects of γ-Radiation* on the Flavour, Texture, Storage and Processing Properties of Fruits and Vegetables*

*Fruit* I—*Strawberries*

Work done in the 1957 season indicated that doses above 0·5 Mrad markedly affected the quality of the berry. Texture and flavour was almost completely destroyed and extensive bleaching was observed concomitantly. These results are in agreement with those obtained elsewhere.[5]

The investigation was resumed this year and the effects of γ-radiation were determined on the incidence of mould attack, flavour, yield and viscosity of expressed juice and ascorbic acid content.

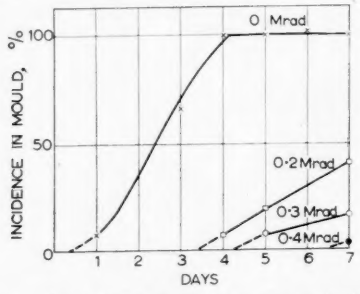


FIG. 1. The effect of γ-radiation on the incidence of mould on strawberries (Cambridge Special) during storage at 20°C.

*Incidence of mould attack.* The incidence of mould attack during storage at 20°C and 1°C was markedly reduced in all irradiated samples. These results are in agreement with those of RAMSEY,[6] VIDAL and BRUNELET.[7] When stored at 20°C, the untreated berries exhibited mould growth within one day, whilst the irradiated samples showed varying periods of delay, related to the amount of radiation they had received in the treatment (Figs. 1 and 2).

It is perhaps noteworthy, that no mould growth was observed on those strawberries stored at 1°C (Fig. 3). Future research should disclose whether the mould spores which can develop at low temperatures have been killed or whether the remaining viable

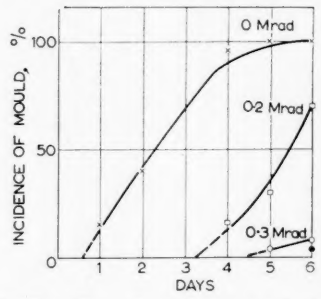


FIG. 2. The effect of γ-radiation on the incidence of mould decay during storage at 20°C on strawberries (Royal Sovereign).

spores cannot recover from some metabolic damage induced by the radiation treatment.

*Flavour.* As the dose was increased from 0·2 to 0·4 Mrad, the panel were able to detect the flavour difference more easily and this is reflected in the increasing value for $X^2$ (Table 1).

Similar results were obtained with another two varieties of strawberries.

*Viscosity and yield of juice.* The viscosity of the expressed juice was reduced from 60 cP in the untreated fruit to 11·2 cP in the irradiated (Table 2). Degradation of pectic substances by ionizing radiation has been known for some time,[8,9,10] and this is the probable cause of the reduction of the viscosity of strawberry juice.

The degradation of pectic substances may

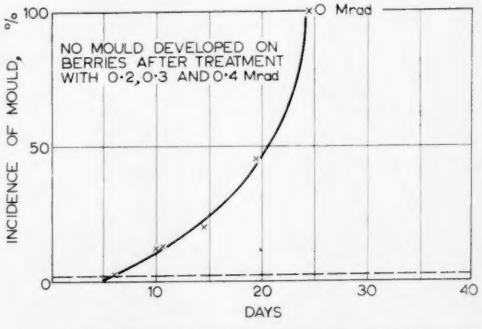


FIG. 3. The effect of γ-radiation on the incidence of mould decay during storage at +1°C on strawberries (Cambridge Special).

* All irradiations have been carried out in air at 20°C.

TABLE 1. The effect of $\gamma$-radiation on the flavour† of strawberries (Cambridge Special)

| Dose (Mrad) | No. of judges* | | $X^2$ | Probability significance |
|---|---|---|---|---|
| | Correct answers | Incorrect answers | | |
| 0·4 | 13 | 6 | 10·53 | 0·01 |
| 0·3 | 12 | 7 | 6·62 | 0·02 |
| 0·2 | 10 | 9 | 3·19 | 0·50 |

*The judges on the panel were selected on the basis of their efficiency in detecting flavour differences.

†Flavour differences were analysed with the aid of the normal triangular test.

also play a part in the induced softening of plant materials,[11,12,13,14] and may also account for a higher yield of juice from irradiated fruit than from untreated fruit (Table 3).

*Ascorbic acid content.* The ascorbic acid content of strawberries was reduced by treatment with $\gamma$-radiation (Table 4).

## II—*Blackcurrants*

The purpose of this investigation was aimed at assessing the potential of the irradiation treatment in the blackcurrant juice industry. Two aspects of fruit processing appeared worthy of investigation. First, the viscosity of the pulp has to be reduced considerably and second, the juice must be sterilized. The effect of $\gamma$-radiation on the flavour of the juice was determined in conjunction with these studies.

*Flavour.* Amounts of $\gamma$-radiation between 0·4 and 0·6 Mrad did not appear to induce large changes in flavour, and, as in the case of strawberries, it proved necessary to select the panel judges on the basis of their efficiency in detecting flavour differences.

TABLE 2. The effect of $\gamma$-radiation on the viscosity of strawberry juice

| Date | Variety | Dose (Mrad) | Viscosity of juice (cP) |
|---|---|---|---|
| 26/6/58 | Royal Sovereign | 0 | 60·0 |
| | | 0·4 | 11·2 |

TABLE 3

| Date | Variety | Dose (Mrad) | Vol. of expressed juice (ml/100 g F.W.) | | % increase |
|---|---|---|---|---|---|
| | | | control* | irradiated* | |
| 26/6/58 | Royal Sovereign | 0·4 | 47 | 58 | 23 |
| 3/7/58 | Cambridge Special | 0·4 | 44·5 | 55 | 22 |
| | | 0·3 | 44·5 | 52 | 15 |
| | | 0·2 | 44·5 | 56 | 24 |

* The mean of duplicates.

It is interesting to note that the judges who were reliable for strawberries were not consistent in their judgements on blackcurrant juice.

By this means, it was possible to demonstrate that flavour changes have been induced at the dose levels employed, but it is believed that these changes are not critical with regard to consumer acceptance. Indeed, some people considered that the flavour of the irradiated juice was preferable to that of the untreated juice.

*Yield and viscosity of expressed juice.* Whole berries were irradiated and the effect of $\gamma$-radiation on the viscosity and yield of juice was determined immediately after irradiation and some hours later by conventional methods (Table 5).

As can be seen from Table 5, the viscosity of the expressed juice from irradiated blackcurrants was very much reduced. Small increases in the yield of juice have been noted and this was probably due to the lowered viscosity of the juice from the irradiated berries.

TABLE 4. The effect of $\gamma$-radiation on the ascorbic acid content* of strawberries (Cambridge Surprise)

| Dose (Mrad) | Ascorbic acid mg/100 g F.W. |
|---|---|
| 0 | 75·0 |
| 0·3 | 28·0 |
| 0·4 | 14·2 |

* Determined by the method of HUGHES.[10]

The present commercial practice is to employ a fungal preparation of polygalacturonase to degrade the pectic substances in the juice, which are the principal compounds causing the high viscosity of the pulp. The effect of $\gamma$-radiation on the viscosity and yield of juice was determined in the presence and in the absence of the enzyme (Table 6).

TABLE 5. The effect of $\gamma$-radiation on the yield and viscosity of expressed juice of blackcurrants (MENDIP)

| Date | Period of storage at ambient temperature after irradiation | Dose (Mrad) | Yield (ml/200g F.W.) | Flow time* (sec) |
|---|---|---|---|---|
| 17/7/58 | 0 | 0 | 50 | 960 |
| | | 0·4 | 59 | 187 |
| 24/7/58 | 0 | 0 | 63 | 536 |
| | | 0·4 | 73 | 378 |
| | 26 | 0 | 60 | 245 |
| | | 0·4 | 65 | 93 |
| 31/7/58 | 0 | 0 | 60 | 1560 |
| | | 0·6 | 64 | 166 |

* Measured in a No. 3 Ostwald-Fenske viscometer at 25°C.

The results indicated that after a period of a few hours the rate of enzymic reduction of viscosity of the irradiated pulp was about identical with that of the untreated pulp. A later experiment (Fig. 4) showed that the differences in the viscosities were only apparent within the first few hours.

The evidence indicated that the enzymic treatment was far superior to radiation alone and that there was no benefit to be derived from combining the two processes. Nevertheless, the manufacturers find difficulty, occasionally, with the enzymic process and it would be interesting to determine the effect of radiation under such circumstances.

*Microbiology.* At dose levels between 0·4 and 0·6 Mrad, the blackcurrants were free

TABLE 6. The effect of $\gamma$-radiation on the viscosity and yield of juice from blackcurrant pulp in the presence and absence of fungal polygalacturonase

| Date | Variety | Period after irradiation (hr) | Treatment | Dose (Mrad) | Viscosity (cS) | Yield ml/200 g F.W. |
|---|---|---|---|---|---|---|
| 24/7/58 | Mendip | 2 | Enzyme and untreated pulp | 0·4 | 1·91 | 115 |
| | | | Enzyme and irradiated pulp | 0·4 | 2·18 | 110 |
| | | | Enzyme added after irradiation | 0·4 | 2·09 | 105 |
| | | 6 | Enzyme and untreated pulp | 0·4 | 1·73 | 138 |
| | | | Enzyme and irradiated pulp | 0·4 | 1·86 | 138 |
| 31/7/58 | Wellington XXX | 3 | Enzyme and untreated pulp | 0·6 | 11·83 | 60 |
| | | | Enzyme and irradiated pulp | 0·6 | 4·55 | 60 |
| | | 8 | Enzyme and untreated pulp | 0·6 | 2·09 | 100 |
| | | | Enzyme and irradiated pulp | 0·6 | 2·45 | 100 |
| 7/8/58 | Baldwin | 6 | Enzyme and untreated pulp | 0·5 | 3·36 | 115 |
| | | | Enzyme and treated pulp | 0·5 | 3·16 | 111 |
| | | 8 | Enzyme and untreated pulp | 0·5 | 2·64 | 136 |
| | | | Enzyme and treated pulp | 0·5 | 2·73 | 130 |

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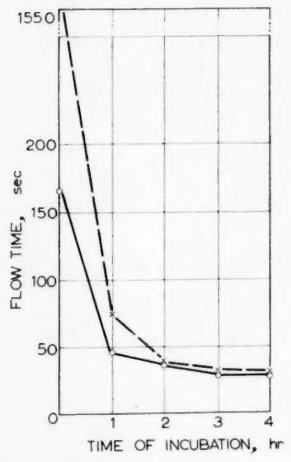


FIG. 4. The effect of $\gamma$-radiation on the rate of reduction of viscosity of blackcurrant pulp by a pectinase preparation.

from a significant number of mould spores, but a considerable number of viable yeast organisms were detected even after treatment with 1 Mrad. Since the problem in the factory is primarily a yeast problem, no immediate application of ionizing radiations can be foreseen in the sterilization of blackcurrant juice.

## III—*Plums*

The amount of information available is much less for plums than for other fruits.[5]

Preliminary results indicated that plums which had received 0·4 Mrad were free from a significant number of mould spores but that viable yeasts though considerably reduced in number, were still present on the surface of the fruit (Table 7).

TABLE 7. The effect of $\gamma$-radiation on the microbial count of plums (Giant Prune) immediately after irradiation and 5 days storage at 20°C

| Storage period (days) | No. of counts/g F.W. of fruit | | | |
|---|---|---|---|---|
| | Untreated | | Treated (0·4 Mrad) | |
| | yeast | moulds | yeast | moulds |
| 0 | 8 | 6 | 15 | 0 |
| 5 | $1{\cdot}7 \times 10^4$ | $2{\cdot}6 \times 10^2$ | $1{\cdot}4 \times 10^2$ | 0 |

## IV—*Apples*

The storage life of apples is dependent upon the mould contamination and also the physiological condition of the fruit.

Very few reports[15,16] have appeared in the literature and the most reliable results can only be described as of a preliminary nature.

Dose levels between 3000 rads and 0·5 Mrad induced softening[10,14] and flavour changes have been detected after treatments of less than 0·1 Mrad. At dose levels between 0·1 and 0·2 Mrad the effect upon texture and flavour did not appear to be too great, but it still remains to be determined whether treatments at these levels will extend the storage life of apples. However some evidence has been obtained that the initial spore count is reduced after treatment with these amounts of radiation (Table 8).

TABLE 8. The effect of $\gamma$-radiation upon the initial microbial count of apples (Cox's Orange Pippin)

| Dose (Mrad) | No. of organisms/g F.W. of peel | |
|---|---|---|
| | moulds | yeasts |
| 0 | $2{\cdot}1 \times 10^4$ | $1{\cdot}4 \times 10^4$ |
| 0·1 | $1{\cdot}1 \times 10^4$ | $1{\cdot}3 \times 10^4$ |
| 0·2 | $3 \times 10^3$ | $3{\cdot}2 \times 10^3$ |

## V—*Pears*

The readily observed effects on pears are very similar to those observed on other fruits, viz. induced softening and loss of flavour.

At dose levels up to 0·55 Mrad, the texture of the irradiated fruit was less firm than that of the untreated and the magnitude of this effect was related to dose (Fig. 5a). During storage at 20°C, the position was eventually reversed and the irradiated fruit was firmer than the untreated fruit (Fig. 5). Similar results were obtained in another five experiments. The delay of softening of pears was accompanied by a delay in the change of colour from green to yellow. Another feature worth mentioning is that at dose levels above 0·4 Mrad, a purple pigment gradually developed after storage for a fortnight at 20°C.

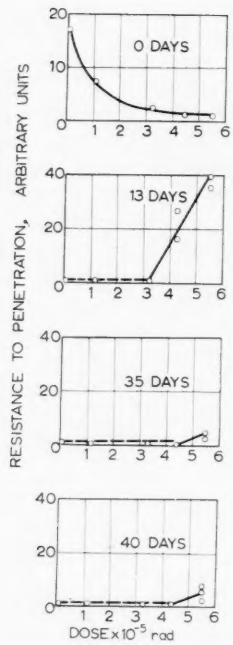


FIG. 5. Effect of $\gamma$-radiation on the texture of Doyenné du Commice pears immediately after irradiation and during storage at 20°C.

It would seem, possibly, that a biochemical mechanism had been induced and such an effect is of interest to physiologists and biochemists.

VI—*Peaches, tomatoes, lemons, oranges, bananas, grapes*

Other workers[6,7] have shown that it is possible to increase the storage life of certain varieties of peaches by at least a factor of 2 at both refrigerated and ambient temperatures with doses between 0·1 and 0·5 Mrad. This effect would appear to be useful in the transport of such fruit.

The ripening of tomatoes has been reported as being delayed by treatment with ionizing radiation.[5,17] This delay of ripening is reflected in a delay of mould attack, but more biochemical analysis is required before it can be established that the ripening as a whole has been affected by this treatment.

Studies with oranges and lemons carried out by RAMSEY *et al.* indicated that storage life at 70–75°C could be appreciably extended but there is the possibility that the fruit would be damaged under present handling conditions.

There is some evidence that the softening of bananas can be delayed[5,16] after treating the fruit with doses less than 50,000 rads, provided that the fruit was irradiated at a suitable stage of maturity. Nevertheless, the advantage in extension of storage life is obviated by damage to the skin.

The quality of grapes appeared to be unimpaired after receiving 0·5 Mrad,[6] which would indicate that radiation may provide a useful control of mould decay in the storage of grapes. Preliminary results obtained at Wantage Radiation Laboratories indicates that after receiving a dose of 0·2 Mrad, the flavour was slightly modified and the texture was less firm but these effects did not appear to be critical in terms of acceptability.

*Vegetables*

There has been no significant advance in this field, as far as it is known, since HANNAN[5] published his own results and reviewed the literature available at that time.

It is generally recognized that a dose of at least 2 Mrads would be required to sterilize the vegetables and it is becoming increasingly clear that the quality of the majority of vegetables is impaired by this treatment. Some work done in America indicates that the refrigerated shelf life of certain blanched vegetables can be almost doubled by doses less than 0·5 Mrad. This would appear to be the most promising line of attack.

## DISCUSSION

*Fresh fruit and vegetables*

In the main, the effect of ionizing radiations on the flavour of uncooked plant materials is to reduce the natural flavour rather than induce the formation of new flavours. Nevertheless, the effects on texture and flavour may prove to be as limiting in the plant industries as the off-odours and

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off-flavours are in the animal product industry.

At the present moment, the evidence indicates that the radiation process is only of a very limited commercial potential, though future research may provide the key which would allow the great, but as yet theoretical, potential of ionizing radiations to be realized fully.

It is well known that enzymic decay can only be slightly modified by comparatively large quantities of $\gamma$-radiation and consequently a combination of refrigeration and irradiation would appear to be the best method for prolonged storage of fresh fruits and vegetables. Also, the results obtained with strawberries would suggest that an increased advantage could be obtained at a low temperature.

All the experiments quoted have been conducted on a small scale and though the results show promise, large scale experiments need to be conducted under simulated commercial practice before conclusions can be drawn as to the advantages of the radiation process.

*Processed fruit and vegetables*

The canning, jam manufacturing, and fruit juice industries are included in this section.

A dose level exceeding 1 Mrad is required to sterilize all plant products and it has already been pointed out that such dose levels normally result in a reduction of quality. Consequently, unless it is shown that micro-organisms can be sensitized to radiation or that varieties of fruits and vegetables can be developed which are less susceptible to radiation damage, it seems unlikely, at the present moment, that irradiation will be able to compete successfully with existing commercial processes. Nevertheless, there is still the possibility that the radiation process may be applied in extending the storage life of the raw product before it is processed in the factory.

*Surface sterilization*

Theoretically, there is no objection to surface radiation in the case of some products, and, indeed, some advantage may be gained by killing the spores on the surface without affecting the remainder of the tissue. Some success has already been noted in the case of peaches[18] but it is not known if this process can be adapted to suit commercial handling of other fruits.

*Acknowledgements*—I wish to acknowledge the technical assistance given by Miss M. Burt and Miss I. Blake and to thank Messrs. H. W. Carter and Sons, Coleford, Glos., for their co-operation in the experiments on blackcurrants.

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# Fresh Fruits and Vegetables

G. TRIPP*

Quartermaster Food and Container Institute for the Armed Forces, Chicago, Ill., U.S.A.

DR Clarke has described the various aspects of fruit and vegetable irradiation quite thoroughly. However, there is one point discussed in his paper and a few not considered which require comment.

The pH of fruits and vegetables has a decided influence on the sterilizing dose required. Inasmuch as *Cl. Botulinum* is not able to grow in an acid medium this organism will not be troublesome in foods having a pH less than 4·6; however, in these circumstances the precise sterilizing dose is not known. When the pH exceeds 4·6 results indicate that a dose of 4·8 Mrads will be required.

In some instances investigators have irradiated fruits and vegetables in sealed plastic bags such as polythene. Items treated at pasteurizing doses continue to respire and this method of packaging can be harmful to the item since normal respiration cannot take place. When reading reports of work it is well to note the type of packaging used since improper packaging can result in undesirable changes far more severe than those induced by radiation.

Irradiation of carbohydrates in aqueous solution results in the formation of hydrogen gas. When fruits and vegetables are radiation sterilized in tin-plate cans, the gas released is sufficient to produce bulged cans. This would be troublesome from the marketing standpoint because bulging of the cans is normally used as a spoilage criterion. This can be circumvented by leaving larger headspace in the can and closing under high vacuums (from the canning viewpoint about 15 to 20 in. of vacuum). Inasmuch as the processor may be required to leave larger headspaces he will also leave more oxygen in the can. Since oxygen plays a role in the quality of the irradiated food it is well to use these conditions in experimental packs of fruits and vegetables.

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* FAO Consultant.

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# Discussion

Mr. VIDAL (France): I agree with the opinions expressed by Mr. Burton and Dr. Mikaelsen, but I would like to comment on a few points. Dr. Burton referred to the difference of dose needed to arrest the germination of potatoes when using different radiation sources. I think that this can be explained by the nature of the radiation source used. When using a 0·5 MeV source a large part of the potatoes would not be irradiated and the low penetration power of electrons from such a source would mean that the eyes of the potatoes would not be completely destroyed. I consider that if machine sources of irradiation are to be used for the suppressing of sprouting of potatoes high level electron energies will have to be used.

In order to obtain the best results in the irradiation of potatoes, it is necessary to consider various factors such as the physical and chemical condition of the potatoes and the conditions of storage (temperature and humidity). For short term storage, doses of 5000 to 6000 rads are sufficient but for long term storage (12 months or longer) it is necessary to use doses of 7500 to 10,000 rads. Since at storage temperatures of +2 to +4°C, the sugar content of potatoes increases, it is advantageous to use as high a storage temperature as possible, while to avoid microbial wastage, the relative humidity should be relatively low, but undue losses due to evaporation should be avoided. As regards time of irradiation after harvest, some of our investigations would indicate that the best results are obtained if irradiation is carried out about a month after harvest.

The results of our investigations on the irradiation of fruits and vegetables are in line with those reported by Dr. Clarke. Temperatures in the range of +4 to +7°C are used for commercial storage and transport of fruits and vegetables, and using radiation doses of 150,000 to 200,000 rads, we have been able to extend the storage life of some fruits and vegetables by a factor of 2 or 3 with little radiation effect on the texture and flavour of the produce.

With regard to tomatoes, we started last year an important programme of work ranging from the irradiation of seeds to obtain increased productivity to irradiation of the tomatoes themselves to increase their storage life. There are indications that the irradiation of green tomatoes with doses of about 500,000 rads followed by storage at about +4°C might considerably increase their storage life. Before using, the tomatoes are given a heat treatment to ripen them.

Dr. DOUZOU (France): We agree with the conclusions of Dr. Burton concerning the reduction of ascorbic acid content in potatoes that have been irradiated and stored.

However, we have been able to show experimentally that the diminution of reducing power of the fractions containing vitamin C does not correspond to a destruction of these fractions, but to a change in their physical–chemical form to dehydro-ascorbic acid. This form is radiation resistant and, under favourable conditions (such as alkaline conditions of pH), can in turn change into compounds which have the same chemical properties and vitamin activity as the ascorbic acid precursor.

Dr. Tripp has told us that the irradiated canned foods liberate hydrogen and oxygen and that, in the case of oxygen, this constitutes an element of danger that cannot be ignored. It should be noted that oxygen constitutes a scoria of the radio-chemical process and the action of the radio-chemical precursors as well as the accompanying long-term effect should be considered. This is a problem which we are at present studying.

Dr. CONTREIRAS (Portugal): I would like to ask Dr. Burton whether or not the marked starch hydrolysis observed in irradiated potatoes was accompanied by a parallel change

in the respiration rate and therefore by a variation in dry matter as compared with controls.

Dr Burton (U.K.). The rise and subsequent fall of sugar content which follows irradiation is accompanied by a rise and fall of the rate of respiration. If irradiation is carried out immediately after harvest, the rate of respiration has, within a month or so, fallen to a level similar to that of the control tubers. We have not measured the respiration during the final sweetening but there is a marked loss of dry matter at this time, and I feel that in all probability respiration may have increased.

Dr. Ericson (Sweden): Reference was made to the work of Dr. M. Jaarma, The Institute for Organic Chemistry and Biochemistry, University of Stockholm, who has studied the influence of ionizing radiation on potato tubers. Dosages in the range of 4000 to 16,000 rads markedly increased the sucrose content and to a lesser degree the glucose content but decreased the fructose content. The changes were reversible and disappeared on storage at +5°C. A dose of 16,000 rads was generally sufficient to cause a nearly complete inhibition of sprouting, but it was found that the dose required for complete inhibition of germination depends on the variety, the dose rate, and the elapsed time after harvest.

Dr. Stosić (Yugoslavia) reported some results obtained in the irradiation of wines in Yugoslavia to control the growth of certain undesirable fungi. Doses required to obtain this control varied greatly and some fungi such as *Botrytis cinera* are able to develop after irradiation at a dose of 0·6 Mrad. Changes in colour, specific gravity and chemical composition of wine were brought about by irradiation and these changes are similar to those that occur in matured wines.

Mr. Vidal (France) reported work on wines, having obtained different results from the Yugoslav workers. Much more experimental work is needed; it is possible to age wines with low doses, and obtain a madeira type wine. It is also possible to change a poor quality wine into a vermouth type.

Dr. Jensen (Denmark): We started with the irradiation of blackcurrants in the past season. The results of the few experiments we have done are very similar to those of Dr. Clarke's. After a dose of 500,000 rads we have obtained the darker colour in juice of the irradiated blackcurrant and we have found that the content of ascorbic acid remains at the same level before and after radiation. Our taste panel on several occasions preferred juice from the irradiated blackcurrants. We are interested in doing more work on this subject and I therefore ask for figures on the colouring in the juice, and for the content of ascorbic acid in Dr. Clarke's experiments.

Dr. Clarke (U.K.): In reply to Mr. Jensen I would like to say that we have not made any quantitative observations on the effect of radiation on the colour of blackcurrant juice, but we did observe an enhancement of the colour after treatment with 400,000 and 500,000 rads. It would appear that the vitamin C content of blackcurrants is not appreciably affected by doses up to 600,000 rads. Perhaps Dr. Uri would be interested in this result.

Dr. Uri (U.K.): I am indeed interested in the resistance of ascorbic acid in irradiated blackcurrant and its juice. Blackcurrant is particularly rich in flavonols and our work on oxidative rancidity indicated that flavonols can be very efficient free radical acceptors. This is certainly an interesting subject for further investigation.

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# The Effect of Ionizing Radiation on Parasites, or Destruction of Trichina in Pork Carcasses

H. F. KRAYBILL*

Chemistry Division, U.S. Army Medical Research and Nutrition Laboratory, Denver 8, Colorado, U.S.A.

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In 1954 GOMBERG *et al.*,[1] at the University of Michigan, established the feasibility of controlling trichinosis by irradiation of pork. About 0·70 to 0·93 Mrad are required to kill trichinae *in situ*, but 18,000 rads of $Co^{60}$ $\gamma$-rays will stop the development of larvae to adults, and 11,000 rads will sterilize the females, thus preventing reproduction. Operating at these low radiation doses is economical, and the amount of irradiation applied to a hog carcass is not enough to produce the deleterious off-odour and off-flavor changes encountered at the sterilization dose of 2·8 Mrads. Mixed fission products and 200 kV X-rays have also been used effectively in this processing of pork.

Using $\gamma$-rays with energies of 0·6 MeV or greater to irradiate the hog carcass, the dose variation was only 15 per cent. With the irradiation source approximately 13 to 17 in. from the hog carcass, the radiation penetration required was 10 to 14 in. At these distances, the dosimetry varied about 2 to 1 from the surface to the interior of a 14-in. thick section.

Using the $Co^{60}$ source the irradiation was conducted in air, while with mixed fission products the irradiation was conducted in a 20-ft deep water canal. In the experimental work conducted by the University of Michigan group, the times for successful treatment of a hog carcass with radiation from mixed fission products varied from 320 min for the hind quarter to 1185 min for the fore quarter. Since a 24-hr period is usually allowed in meat packing plants from the time a hog is eviscerated and then cooled prior to cutting, this elapsed time was used profitably by the irradiation treatment of the carcass.

To test the biological effectiveness of this process in the destruction of encysted trichina larvae, feeding experiments were conducted with rats. Five thousand larvae which had been recovered from a sample of irradiated meat were fed by stomach tube to a group of rats. The control rats received 5000 larvae from non-irradiated meat. Examination of the small intestine of these rats was made 6 days after feeding. Thirty days later the muscles of these animals were examined for evidence of encysted larvae. Using the cobalt-60 source, it was found that a dose of *11,000 rads was required* for complete sterilization of the female trichinae, whereas with the mixed fission product source complete sterilization of females appeared to be accomplished at *9300* rads.

The irradiation of the pork has the added advantage of greater refrigerator shelf life as determined by taste panel tests. With irradiation doses up to 93,000 rads, no differences in flavor were detected by a taste panel when comparing the treated product with unirradiated pork.

This group of investigators has calculated the cost of this operation based on a plant that has an output of 2000 hog carcasses per day. These calculated costs included interest on investment with allowance for five-year amortization period. The irradiation cost per carcass would be approximately 2·3 mills or 0·23 of a cent per pound for a 135-lb finished carcass (excluding lard).

The importance of this process as a public

* FAO Consultant on microbiology. Present address: Curtiss Wright Corporation, Princeton, New Jersey, U.S.A.

health measure is readily appreciated. Despite statements to the contrary, a significant segment of the population consumes raw pork, causing trichinosis to be a public health problem. Convincing evidence for this statement has been provided by pathologists who have reported encysted trichinae in 16 per cent of post-mortem examinations.

In the United States severe control measures have been proposed for elimination of trichinae and ultimately trichinosis:

(a)[1] Legislation by 47 of 49 states requiring the cooking of garbage fed to hogs.

(b) By freezing hog carcasses as follows:
5°F (20 days) −10°F (10 days)
−20°F (6 days) and −36°F (2 min)

(c) By smoking meats to an internal temperature of 137°F.

(d) By the consumer cooking pork until meat turns grey or grey–white color or until internal temperature of meat has exceeded 137°F.

Again irradiation of hog carcasses at low radiation doses should provide an absolute safeguard for controlling this parasite.

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# The Effect of Ionizing Radiation on Insects

H. F. KRAYBILL*

Chemistry Division, U.S. Army Medical Research and Nutrition Laboratory, Denver 8, Colorado, U.S.A.

SEVERAL methods have been used for elimination of insects from grains and foods. The alternative methods are as follows:

(a) High frequency heating.
(b) Sonic vibration.
(c) Chemical fumigation.

The fumigation has several disadvantages. The hydrocyanic gas used is toxic to humans, has a concentration which persists in product for a long time and this fumigant cannot penetrate pocket.

The use of ionizing radiation usually requires doses of 25,000–50,000 rads to destroy insect eggs and 300,000–600,000 rads will be lethal to all insects. The radiation dose of 25,000 rads prevents development of insects from one metamorphic stage to the next and also prevents reproduction in the female. The species of insects studied are as follows:

(a) Confused flour beetle.
(b) Yellow mealworm.
(c) Sawtooth grain beetle.
(d) Lesser grain borer.
(e) Cigarette beetle.

The radiation dose selected applies equally to all insect species. The determining factor in application of radiation is the effect on quality of product. It may be well to indicate the effect of radiation dose on some cereal products.

*Wheat*

250,000–500,000 rads applied will produce a satisfactory loaf of bread with only slight objectionable flavor.

*Flour*

The off-flavor threshold on this product occurs at 20,000 to 50,000 rads. At this radiation dose a satisfactory bread and cake was obtained with slightly reduced volume for the cake.

*Oats*

At 50,000 to 1,000,000 rads no effect on oat stability or nutritive value was noted. No material difference other than slight flavor change.

*Spices*

Treating cinnamon at 75,000 rads yielded a satisfactory product but increasing dosage to 150,000 rads produced a bitter flavor. The other spices treated such as paprika, ginger, aniseed, nutmeg, Chili powder and coriander were satisfactory products at this radiation level.

A study has been carried out by the United States Department of Agriculture in which the screw worm fly has been treated with ionizing radiation. The screw worm fly plagues animals in the southeastern portion of the United States causing not only flesh wounds and crippling the animal through muscle damage, but results in hide or leather damage. The cost of treating cattle in Florida alone has been estimated at $20,000,000 per annum.

The female fly lays masses of 200 or more eggs on the edge of cuts, wounds, sores and scratches on cattle. In 3 days these eggs hatch into larvae which feed on the living flesh. By irradiating adult male flies they become sterile. These males only mate once with females and since they are sterile the reproduction cycle is interrupted.

It is hoped that this method of control can be applied to other disease carrying insects such as the blowfly and the African tsetse fly. The effectiveness of radiation in producing sterile males or females largely depends on the proportion of insect population that can be treated and released for mating and the frequency with which the irradiated insects undergo mating.

* FAO Consultant on microbiology. Present address: Curtiss Wright Corporation, Princeton, New Jersey, U.S.A.

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# The Disinfestation of Foods, particularly Grain

P. B. CORNWELL

Technological Irradiation Group, Isotope Division, Wantage Radiation Laboratory, Wantage, Berks

There are two potential methods of controlling insect pests by irradiation; indirect control by the release of sterilized adults and the direct treatment of infested products.

Application of the first method is limited to certain species by a number of biological and environmental factors. The second method could have wider application for the control of all species infesting stored materials. Among the factors which may influence its success, problems associated with bulk handling and the incorporation of treatment into established handling procedures, must receive primary consideration. These could be most easily solved in the treatment of packaged goods where production and irradiation may be carefully integrated.

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Insects attack foodstuffs at various stages of production between the growing crop and the retailed product. Thus, standing maize may be attacked by weevils (*C. oryzae*) which fly into the field and infest the ripe cobs before harvest. Frequently, products in store become infested by the spread of a pest population resident in the storage premises. A further site of infestation may be in the processing factory, giving rise to the export of infested, packaged goods. Established trade channels have resulted in the transport of pests from one country to another, such that produce may become infested during shipment. Finally, insect attack may occur on the shelf in the retailer's shop or in the domestic larder. The greatest losses in weight, quality and monetary value occur when foodstuffs are retained in storage for considerable periods, particularly under tropical and sub-tropical conditions.

Recognition of the growing resistance of insects to conventional insecticides has given greater impetus in recent years to the search for more effective methods of insect control. Research into the potentialities of ionizing radiations has been stimulated by the advent of large sources of $\gamma$-radiation which utilize the by-products of the nuclear power industry. A number of independent studies on the effects of $\beta$, $\gamma$- and X-rays on insect pests show that all three radiations are comparable in their lethal and sterilizing properties.[1,2] In this review, the fundamental and applied problems of using large sources of $\gamma$-radiation will be considered.

## METHODS OF RADIATION CONTROL

Two techniques for radiation disinfestation can be distinguished: the direct irradiation of infested products,[3] and the indirect control of a pest species by the release of sterilized adults.[4] The first method involves the irradiation of insects with the products which contain them to kill or induce permanent reproductive sterilization. The second method involves rearing vast numbers of the pest species, irradiating the pupae and allowing the sterile adults to emerge within the environment of the pest. This technique is of limited application and will be considered first.

### *Indirect control by the release of sterilized adults*

Control by this method is achieved by swamping a pest population with sterile

FIG. 1. Hypothetical figures illustrating the principle of reducing an insect population by the sustained release of sterile males

| Natural population of females | No. of sterile males released per generation | Ratio of sterile to fertile males | Percentage of females mated to sterile males | Theoretical population of fertile females each subsequent generation |
|---|---|---|---|---|
| 1,000,000 | 2,000,000 | 2 : 1 | 66·7 | 333,333 |
| 333,333 | 2,000,000 | 6 : 1 | 85·7 | 47,619 |
| 47,619 | 2,000,000 | 42 : 1 | 97·7 | 1,107 |
| 1,107 | 2,000,000 | 1,807 : 1 | 99·95 | Less than 1 |

Reprinted from a paper by KNIPLING E. F. in *J. Econ. Ent.* **48,** 459 (1955).

insects, such that the reduction in numbers and final eradication of the species is obtained by the competitive mating over a number of generations of wild and sterilized males. The treatment could be applied to the control of insects in the field and in manufacturing premises. However, the requirements for successful operation of the method[5] indicate that it could be applied only to a limited number of species:

(i) that the female of the pest species mates only once,
(ii) a mass rearing technique is available to produce the vast numbers of insects required,
(iii) the pest population is inherently low or can be conveniently reduced to a low level by conventional insecticides,
(iv) the species exists in a closed environment,
(v) the adult stage is non-destructive,
(vi) the released insects can be distributed easily,
(vii) the insects can be adequately sterilized, without adversely affecting their mobility, mating behaviour or reducing their length of life.

For the majority of agricultural pests it would be extremely difficult to rear the large numbers of insects required. Moreover, their environment frequently extends over many acres or square miles and it would be difficult to prevent the ingress of fertile insects into the release area. However, the method could, perhaps, be applied where a closed environment is provided by the four walls of manufacturing premises, particularly of flour mills. The control of flour mill moth by this method is receiving intensive study at theWantage Radiation Laboratory; *Ephestia kühniella* can be reared in vast numbers, and by releasing sterile moths into infested mills it may be possible to eliminate completely the population for a number of years. The pest population could be reduced in the first instance by annual fumigation, but there is some evidence to doubt whether a flour mill really constitutes a closed environment, for moths may be reintroduced into the premises on infested sacks or with the incoming grain. It is obviously impossible to assess the cost of such a treatment at this stage.

*Direct treatment of infested products*

The control of pests by direct treatment has far greater potentialities than the first method. It is most suited to the disinfestation of products handled in bulk at particular centres. Whilst this is a feature of many foodstuffs, the method is particularly suited to the treatment of cereals, and for this reason the effects of $\gamma$-radiation on cereal pests have been studied in detail. However, irradiation offers no protection against reinfestation and is therefore most effective when followed by storage in silo bins which, together with boat holds, could conceivably be freed from resident infestations. Without the construction of insect-proofed warehouses it is difficult to visualize that irradiation could be of benefit for the treatment of produce in sacks or open boxes. The most useful application could be for the control of

insects in packaged foods which are safeguarded against reinfestation by an insect-proof wrapper.

(a) *Dose level for treatment.* Insect control could be achieved by one of three dose levels.

(i) Doses of about 0·5 Mrad to provide reproductive sterilization and death within 24 hr.[6,7] This level provides a high measure of control of the microflora[8] and could allow grain storage at higher moisture contents.

(ii) Doses of about 100,000 rads to provide sterilization and death within 7 days; to induce radiation lethargy[9] so that further damage caused by the insects feeding before death is minimal. This level would sterilize mites[10] but provide no control of microfloral activity.

(iii) Doses of 15–20,000 rads to provide sterilization and death of insects in about 3 weeks.[11] Adults continue to feed during the survival period; the development of immature stages is arrested. These doses would not reduce the population of mites.

On economic grounds only the lowest dose range can be considered of immediate interest.

(b) *Radiation susceptibility of species, developmental stages and strains.* Cereals support a variety of insect species and a dose evaluated for commercial treatment must be effective against the most resistant. Studies on 13 species infesting grain indicate that they are comparable in susceptibility to sterilization, but they differ markedly in their lethal response.[12]

Radiation susceptibility varies with the developmental stage of the insect. Thus, adult grain weevils fail to develop when eggs and larvae are treated with doses above 5000 rads, but 20 per cent of irradiated pupae may produce adults when treated with 20,000 rads. The latter dose does not prevent the emergence of adults preformed in the grain. All stages of the grain weevil can be sterilized by doses of 15–20,000 rads, this

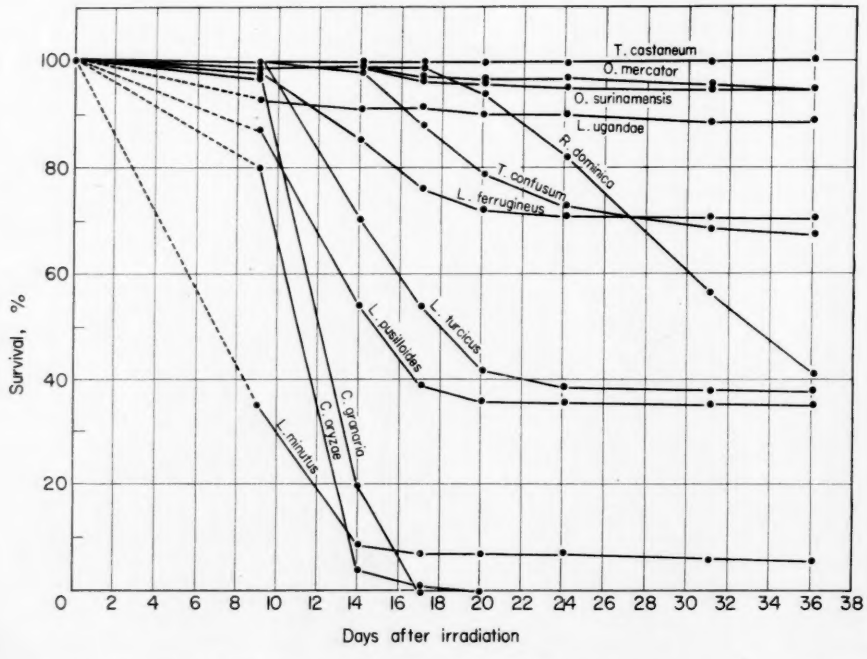


FIG. 1. Survival of adults after treatment with $6 \times 10^3$ rep $\gamma$-radiation. Data corrected for mortality in the controls. (Reproduced from *Nature* **179,** 670 (1957).)

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upper dose level allowing a considerable safety factor for commercial treatment.[11]

Since irradiation could be applied to control insects originating in various parts of the world, differences in strain susceptibility must be considered. Tests on a number of strains of *C. granaria* have shown considerable variation in the lethal response but differences in the degree of sterilization are slight.

*Fundamental problems of using radiation treatment*

One of the most important problems which should be considered under this heading concerns the possible development of radiation resistance. In practice, the use of sub-sterilizing doses could result in the production of progeny which require a higher dose to kill or sterilize. Resistance to conventional insecticides can develop by the process of selection. Since one effect of radiation is to produce mutations, resistance may develop even more readily through the combined effects of selection and these genetic changes. A recognition of this possibility must be reflected in the provision of an adequate "safety factor" in evaluating doses for commercial treatment.

When using sub-sterilizing doses, most of the fertile eggs are produced during the first few days of the survival period. Current studies at Wantage[13] are directed towards increasing radiation susceptibility to obtain a more immediate kill. A faster rate of mortality might be obtained by combining irradiation with other treatments, e.g. different temperatures or concentrations of different gases. Success in this direction could lead to a lowering of the dose level required for commercial treatment. Using the grain weevil in studies with fractionated doses it has been shown that the efficacy of direct irradiation could be seriously jeopardized if these treatments were separated by intervals of more than a few minutes.[14] A recognition of this problem must be reflected in the construction of irradiation plant of sufficient capacity to make multiple treatments unnecessary.

We should also consider possible detrimental effects of irradiation on the food itself —changes in nutritional value, baking quality, flavour and colour, affecting consumer acceptability. Reports in the literature indicate that doses of less than 100,000 rads do not cause measurable changes in quality. However, workers have shown loss of odour and flavour in breakfast cereal treated with 60,000 rads[15] but no changes in the quality of cake mixes irradiated at this level. The quality of bread produced from hard red winter wheat irradiated at 1 Mrad is described as acceptable, and changes in the biochemical factors and dough properties caused by doses up to 0·6 Mrad are reported as not damaging to breadmaking quality.[16] Current work at Wantage has shown that the treatment of English wheat with 0·5 Mrad produces slight changes in baking quality which are of no commercial significance.[17]

These reports confirm the absence of changes in quality with sterilizing doses, but it is emphasized that each product must be individually tested. Provision must also be made for examining changes in nutritional value.

*The design of a γ-facility for disinfesting grain*

Russian literature describes a semi-production type $Co^{60}$ facility with an output of 1·85 tons/hr. The output of plant containing 3·72 Mc of fission products is given as 31 tons/hr. American workers describe an irradiation chamber for treating flour at 27 tons/hr.[18]

At present, it would be correct to say that we have only first ideas on the major principles involved in designing an irradiation facility for this purpose. Among the many factors which may influence the successful application of radiation disinfestation, the incorporation of treatment into established handling procedures is of fundamental importance.

Two types of grain conveyor are in current use; the slow moving spiral worm and the pneumatic plant carrying grain with air speeds of 100 miles/hr. The density of moving grain in the pneumatic conveyor is about 1/60 of the static bulk density. Thus, with a 10 cm tube (a typical diameter of pneumatic plant) the radiation utilization is

about 0·5 per cent and 99·5 per cent of useful radiation is wasted outside the operating field. To improve the utilization to 25 per cent, a tube of some 7 m radius is required and to handle useful quantities of grain the source would have to be long and thus involve large shielding problems. This could be overcome by underground handling, but at present such systems have been developed only in exceptional cases to suit circumstances where operation above ground is impossible. Accordingly, pneumatic conveyance during irradiation holds little promise.

With a slow moving worm conveyor, 1 Mc of $Cs^{137}$ could deliver 1 Mrad to 700 lb of grain/hr using 25 per cent of the available radiation. If this source is enclosed in an 8 in. diameter cylinder 10 ft long, and surrounded by an outer cylinder of 2 ft diameter, the capacity of the 8 in. wide annulus is 28 $ft^3$ and would hold 0·56 tons of grain. Using a flow rate of 5 ft/min, a dose of 20,000 rads could be delivered to 17 tons of grain/hr. This arrangement, holding static grain, would provide a difference in the dose rate of $\times 3$ across the annulus. Good mixing during irradiation is essential; uniform movement in the feed could be assisted by a vertical flow, and this arrangement would also favour the loading of sources and their removal to a safe position. Five feet of concrete would be required for adequate shielding.

This basic design can be improved by surrounding the source with two more cylinders to provide further annuli of 56 and 84 $ft^3$ capacity. The grain could be fed at different rates, first through the outer annulus and then through the inner ones. This arrangement increases the radiation utilization to 40 per cent and the overall handling rate to 25–30 tons/hr.

The same dose and handling rates could also be obtained by using 210,000 c of $Co^{60}$, but this isotope decays five times more rapidly than $Cs^{137}$ and the cost of treatment would be very similar. Either source would be allowed to decay by 10 per cent before replenishing with more active material, this taking place every 10 months for cobalt and every 4 yr for caesium.

The prices of cobalt and caesium in 1963 may be estimated at 5s. and 10s. per curie, respectively, followed by a five-fold reduction by 1968. In considering all the factors which contribute to total cost, e.g. source decay, depreciation of the specific activity, interest on capital, labour, maintenance, etc., grain could be disinfested by continuous treatment at 1s. 3d. to 2s. 6d./ton in 1963, and at 5s. to 10s./ton when operating only a 40-hr week. The corresponding figures are 3d. to 6d. for continuous treatment and 1s. to 2s. for a 40-hr week in 1968.

The 1963 estimate for disinfesting grain by continuous treatment compares not unfavourably with present-day fumigation costs (1s. to 2s. 6d./ton). The cost of operating a 40-hr week would be acceptable in 1968.

*Applied problems of using radiation treatment*

Insect damage to grain assumes greatest proportions in the areas of production, particularly in the Argentine, East Africa and Australia. It is in these countries that radiation sources could be most usefully installed. Ideally, cereals would be treated before going into storage, the source being located between coming transport and the silo. Treatment could also be applied at the output silo before discharging into boat holds. Imports of grain into Britain are usually milled within a few weeks of arrival and it seems doubtful whether radiation treatment would be of useful application here. However, this does not apply to products such as dried fruits, which are normally fumigated at import.

In the overseas silos, fumigation is operated once or twice a year at fixed points within the bins. At present, this is considered reasonably successful and it is not necessary to reduce further the level of insect population. This position will probably change through the introduction of purity standards (4 insect fragments/lb of flour), already in existence in the United States, and which may well be considered desirable in the U.K. $\gamma$-Radiation can provide 100 per cent control and it is this factor which may promote the use of radiation disinfestation.

Commercial handling rates of grain vary

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from 10 to 100 tons/hr. The capacity of the source described is at the lower end of this range. A throughput of 27 tons/hr could accommodate the intake of grain into silo, but the source would operate at maximum efficiency only during the period of crop production. On the other hand, this capacity would be inadequate for the treatment of grain at the dock side. An average cargo consists of 10,000 tons, for which a number of pneumatic plants would be used, each discharging at 100 tons/hr, completing the operation within 2 days. With a megacurie source operating continuously, the cargo would take more than 2 weeks to discharge. This delay illustrates that the source capacity is of vital importance. It could be increased by housing many megacuries within a common shield, but must provide a minimum throughput of at least 200 tons/hr.

It is felt in certain quarters that irradiation processing could be of considerable benefit to under-developed countries. However, radiation treatment is incompatible with the peasant methods of small-scale storage. The process can be best applied at quaysides or central depots, where trade channels are firmly established, but it is still doubtful whether an increase in export trade, through improved methods of pest control, would ever raise the living standards of a native population. Of real benefit would be increased production by improved methods of cultural practice.

The greatest problems, of applying radiation treatment to agricultural produce for export, are associated with seasonal and annual changes in crop production. The efficient use of an irradiation facility necessitates 24-hr treatment, and shift labour would be involved. At ports of intake, the problem becomes more acute where off-loading may take place along many miles of quayside. Here is a need for a mobile source, sufficiently versatile to treat a variety of products and to accommodate different methods of handling procedure.

On the other hand, the problems of disinfesting packaged goods as they leave a processing factory are comparatively small when cartons are of standard size and shape. With production and irradiation carefully integrated, disinfestation can be applied most efficiently. Nevertheless, it is difficult to cost the value of this process against loss of trade and goodwill.

Our final problem involves public opinion. The public have been made sufficiently conscious of the hazards associated with radiation, that there may be considerable reaction to the treatment of foods. The controversy which has arisen in recent years over the inclusion of various chemicals during bread-making serves as an example. Irradiation processing must be introduced with appropriate steps to ensure public safety and to educate the public of its benefits to mankind.

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# Destruction by Irradiation of Parasites Transmitted to Man through Butchers' Meat

E. L. TAYLOR and J. W. PARFITT
Central Veterinary Laboratory, Weybridge

Meat inspection methods have greatly reduced the risk of the transmission of parasites to man through the consumption of butchers' meat; three parasitic worms still remain, however, in developed countries, *Cysticercus bovis*, *Echinococcus granulosus and Trichinella spiralis*. The incidence of these three has reached a low level but present methods are inadequate to reduce it further.

Observations made at Harwell and Weybridge show that doses up to 100 krads do not kill the larvae of *Trichinella*, nor do they kill *Cysticercus*. These immature stages are, however, rendered incapable of development when transferred to suitable hosts, the inhibitory effects being observable at exposures as low as 10 krads.

It is concluded that the development of suitable methods for applying the treatment coupled with the increased centralization of slaughter might bring about the eradication of *C. bovis* and *T. spiralis*, although *E. granulosus* is likely to remain for a longer period.

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THE continued presence of parasitic worms even among the *élite* of Western civilization shows that the hygiene of our modern cities and homes has reached only a relatively advanced position. Three of these parasites are transmitted through meat, two tapeworms, *Cysticercus bovis* and *Echinococcus granulosus*, and a small roundworm, *Trichinella spiralis*, and are still of frequent occurrence in Great Britain, in common with other developed countries. *C. bovis* is the most common and *Trichinella spiralis* the least common. Another tapeworm, *Cysticercus cellulosae* which formerly was of frequent occurrence in this country now appears to have been exterminated, although it is still present in many other developed countries.

Reasons for the continued presence of *C. bovis*, which is transmitted through beef, and the disappearance of *C. cellulosae*, transmitted through pork, appear to lie in the different habits and husbandry practices concerning these two kinds of animal. Pigs, being scavengers, look for and eat faecal material which may sometimes contain segments of the mature tapeworm. When infection is acquired in this way it is almost invariably a heavy one such as is easily observable during meat inspection and leads to the condemnation of the carcass. Cattle, on the other hand, during the course of their grazing are remarkably careful to avoid all faeces-contaminated herbage; the infestation acquired is therefore light, and as in addition, the carcasses of these animals are large the cysts often escape detection by the meat inspectors. Support for this view was provided by SILVERMAN[1] who in work on this problem showed that birds, notably gulls, feeding on sewage farms are capable of passing the eggs of the worm, still in a viable and infective state, on to the surface of pasture land.

*Echinococcus*, also known as hydatid cyst, is more easily seen than either of these parasites during meat inspection. Its continued presence in our country, however, depends upon the feeding of infective offals to dogs, much of which material has doubtless already been condemned by the food inspectors, and until the malpractice of allowing dogs to eat this material can be stopped the parasite is likely to remain with us.

The position of *Trichinella spiralis* is of another kind in that a reservoir exists in communities of rats which maintain the presence of the parasite through cannibalism,

the infection occasionally passing over to pigs by the eating of rats that happen to be carrying the parasite; it is then easily possible for it to be carried to man. Some 20 years ago HALL[2] made the surprising observation in the United States that up to 27 per cent of the human population there were carrying the infection. The large number of examinations which he carried out *post mortem* in several parts of the country showed that some 16 per cent of the population were infected. A similar search made in our own country following the epidemic at Wolverhampton in which some 500 people were involved, showed the existence of the parasite in 51 out of 472 post-mortem examinations carried out in the mortuaries of hospitals in the region of a number of large towns.[3] Although heavily infected pigs' carcasses may be detected during the course of meat inspection those that are lightly infected are apt to escape and it seems certain that eradication will never be achieved through present meat inspection methods.

The success of an eradication campaign that endeavours to achieve its objective by means that are barely adequate for the task tends to follow the course shown in Fig. 1. A 90 per cent efficacy may bring about a remarkable change in the early phases but as the diminishing few are not completely eliminated for many years it seems needless (to the operatives engaged in the work) to continue the same strenuous efforts that were needed in the first place and the population of parasites tends to build up once more.

The upper part of Fig. 2 shows the position

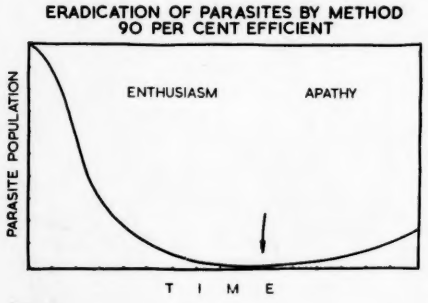


FIG. 1.

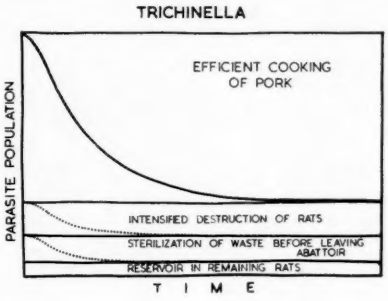


FIG. 2.

for *Trichinella spiralis*, which has been reduced to its present low level of population, partly by the energies of the meat inspectors but principally through the housewife's efficient cooking of pig flesh. It seems unlikely that our present methods will ever succeed in bringing about eradication.

If, however, it were possible to introduce some new measure such as an intensified effort for the destruction of rats, we could, perhaps, proceed one more step towards complete elimination, as suggested by the dotted curve in a lower part of the figure. Enforcement of the sterilization of waste products before they leave the abattoir might have a similar effect but we should still have to contend with occasional transfer from the reservoir of infection in the remaining rat population and could not hope entirely to eliminate all danger of trichinosis until rats had been exterminated—*unless* some new measure is introduced, as for instance the sterilization of all carcasses before the meat leaves the abattoir.

Hydatid cyst is dealt with in a similar way in Fig. 3. In this instance the reservoir of infection occurs in mountain sheep, the carcasses of which are not infrequently eaten by farm dogs or by foxes, which may also disseminate the eggs about the ground. This reservoir of infection is beyond our control at the present time but much could be done through the better inspection of rural slaughter, and the sterilization of all waste material before it leaves the abattoir, which steps would bring about a reduction of the level of infestation as shown in the lower part of Fig. 3.

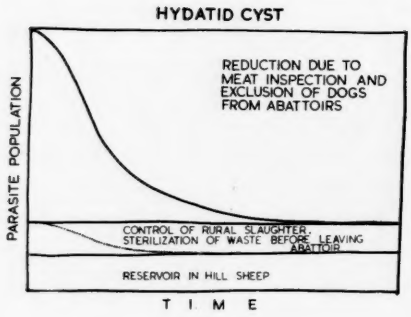
HYDATID CYST
REDUCTION DUE TO MEAT INSPECTION AND EXCLUSION OF DOGS FROM ABATTOIRS
PARASITE POPULATION
CONTROL OF RURAL SLAUGHTER. STERILIZATION OF WASTE BEFORE LEAVING ABATTOIR
RESERVOIR IN HILL SHEEP
T I M E

FIG. 3.

*Cysticercus bovis* is in a different position in that the reservoir of infection is in man himself. The large tapeworm *Taenia saginata* which is the form found in man is very much more prevalent than we like to admit and has an inconveniently long life. Its eggs, which are produced in millions, find their way to the pastures through imperfections in our sewage disposal systems and through dissemination by birds, in particular gulls, which frequent the sewage farms, eat the tapeworm segments that they find there and are able to pass viable eggs through their intestines on to the pastures. It is doubtless as a result of this sparse dissemination of infected material that grazing cattle as a rule carry only a few cysts which, on account of the relatively large size of the bovine carcass, not infrequently escape detection by the meat inspector. When the occasional cyst is found on examination of a carcass, it is an indication of the presumptive presence of other cysts; the small number, however, appears to be inadequate for the total condemnation of the whole carcass which is sterilized by being held at a temperature of — 7°C for a period of at least 3 weeks. This long storage is an expensive procedure in itself, and in addition detracts from the value of the carcass for meat.

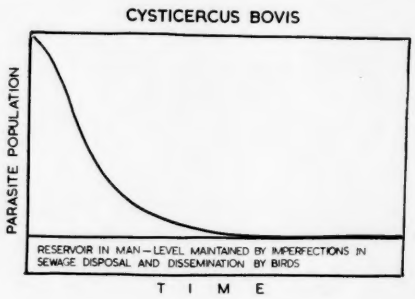
CYSTICERCUS BOVIS
PARASITE POPULATION
RESERVOIR IN MAN — LEVEL MAINTAINED BY IMPERFECTIONS IN SEWAGE DISPOSAL AND DISSEMINATION BY BIRDS
T I M E

FIG. 4.

## EXPERIMENTAL OBSERVATIONS

TABLE 1. Effect of irradiation on parasites in meat

| *Cysticercus bovis* | | | *Trichinella spiralis* | |
|---|---|---|---|---|
| | | | Viability of larvae | |
| Degree of irradiation (krad) | Total cysts | Percentage viable | Muscle digest | Infectivity to guinea pigs |
| Lab. control | 332 | 56·6 | +++ | +++ |
| Harwell control | 253 | 16·2 | +++ | +++ |
| 10 | 334 | 26·3 | +++ | — |
| 20 | 298 | 38·6 | +++ | — |
| 50 | 282 | 25·5 | +++ | — |
| 75 | 287 | 39·7 | +++ | — |
| 100 | 289 | 14·9 | +++ | — |

TABLE 2. Effect of irradiation on *Cysticercus pisiformis*

| Degree of irradiation (krad) | Viability test on 10 cysts Number evaginated | Infectivity test Number of worms recovered from 30 cysts fed to dogs | Size of worms in cm | Day after infection when autopsied |
|---|---|---|---|---|
| Control | 10 | 23 mature | 18·5–36·0 | 42 |
| 10 | 10 | 27 sterile | 6·0–16·0 | 42 |
| 20 | 10 | 16 sterile | 1·7–13·5 | 42 |
| 30 | 10 | 0 | — | 42 |
| 50 | 10 | 21 immature | 0·3–1·5 | 11 |
| 100 | 10 | 0 | — | 42 |

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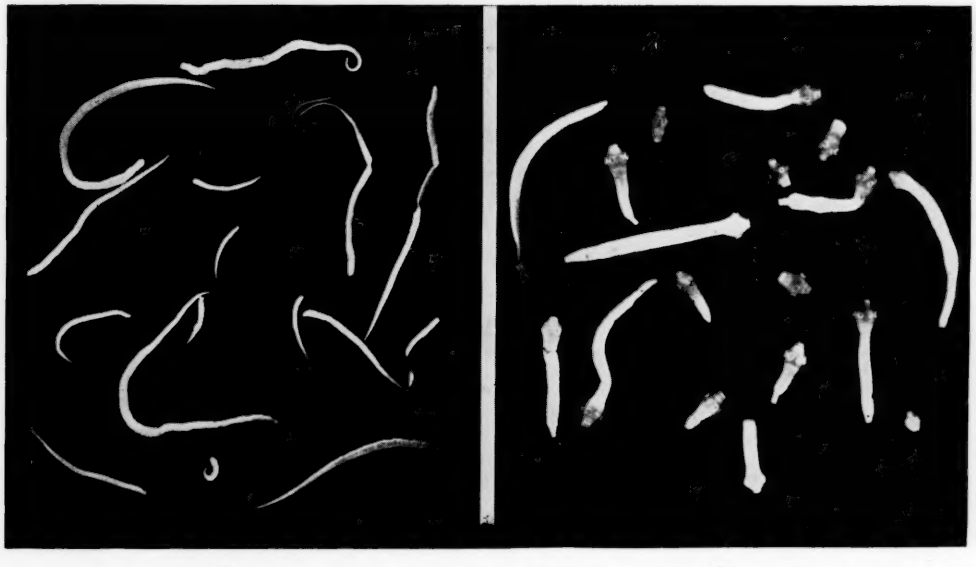
(a) (b)



(c) (d)

FIG. 5(a). Normal *Taenia pisiformis*, $\frac{1}{3}$ actual size, 42 days after feeding of the cysts to a dog.
(b). Abortive *T. pisiformis*, $\frac{1}{3}$ actual size, 42 days after feeding to a dog of cysts exposed to 10 krad $\gamma$-irradiation.
(c). Abortive *T. pisiformis*, $\frac{1}{3}$ actual size, 42 days after feeding to a dog of cysts exposed to 20 krad $\gamma$-irradiation.
(d). Abortive *T. pisiformis*, $2\frac{1}{3}$ times actual size, 11 days after feeding to a dog of cysts exposed to 50 krad $\gamma$-irradiation.

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A young bovine animal experimentally infected with large numbers of *Cysticercus bovis*, was killed at a time when these cysts had reached maturity. Portions of muscle were then dissected away from the bones, taken on the same day or the following day to Harwell and exposed to the various doses of $\gamma$-irradiation from $Co^{60}$. *Trichinella spiralis* in guinea pig muscle, was included as an experimental control to the observations, something already being known of the action of irradiation on these parasites as a result of American work.[4–11]

The particulars of the treatment applied and results obtained are shown in Table 1.

Having confirmed the American observations that although the larvae of *Trichinella* were still viable and that after the treatment they were no longer able to reproduce it was thought desirable to test the same point in *Cysticercus bovis*, which had similarly proved to be alive after irradiation with intensities ranging up to 100 krads. Unfortunately *C. bovis* will not mature in any host other than man, so that it was thought desirable to be satisfied with colateral observations made on a closely related *Cysticercus* in the rabbit, *C. pisiformis*, the mature form of which develops in the dog. The results are summarized in Table 2.

## DISCUSSION

The diverse figures concerning the percentage of viable cysts and appearing in Table 1 are probably to be explained by variations in the vitality of the cysts themselves, which in *C. bovis*, begin to die and degenerate at an early stage after infection has taken place. It is not unlikely that the cysts in the various portions of muscle used for different durations of irradiation contained a variable proportion of dead cysts. This circumstance, however, does not affect the validity of the observations which depend upon the demonstration that cysts which had been exposed to the intensities of irradiation ranging up to 100 krads were still capable of evagination when placed in a solution of bile salts. The point still remaining to be ascertained at that stage in the observations was the infectivity of these cysts—could they grow to maturity in another host, or was their further development completely inhibited as in the example of *Trichinella spiralis*?

The cysts of *C. pisiformis* are much more satisfactory as experimental material than are those of *C. bovis* and figures in column 2 of the table show that 100 per cent of the cysts were capable of evagination in the bile salts. Subsequent feeding to young dogs, which were killed at the end of the sixth week of infection, showed normal mature worms to be present only in the animal which had received thirty normal unexposed cysts. All of the tapeworms which had developed from irradiated cysts were more or less severely affected; the animal which had received thirty cysts exposed to 10 krads was found to contain 27 sterile worms; of the thirty cysts exposed to 20 krads, 16 were found to have made an even less satisfactory growth in the final host, and an irradiation intensity of 30 krads and over, rendered the cysts incapable of growth. A very interesting result, however, was obtained through the experimental dog which received cysts irradiated with 50 krads. This animal had to be destroyed at the eleventh day of infection because of an attack of hysteria, when at the following post-mortem examination 21 minute worms were found to be present. This finding suggested that early examination of the other two animals might have shown the same abortive form of worms to be present at the eleventh day. Photographs of the affected worms are reproduced in Fig. 5.

It may be concluded, therefore, that $\gamma$-irradiation of carcasses intended for human consumption is effective in preventing normal growth and reproduction not only of *Trichinella spiralis* but also of *Cysticercus bovis*. If properly organized it would presumably be less expensive than the cold storage method at present used in Great Britain, against *C. bovis*, and in some other countries against *T. spiralis* as well. One main difficulty would be the centralization of the

slaughter of food animals to such an extent that all carcasses could be treated. That ideal state is unlikely to be reached for a long time; the provision for radioactive treatment at existing large abattoirs or meat storage depots even under our present system would doubtless produce a marked decrease in the incidence and go a long way towards the eradication of *Cysticercus bovis* and *Trichinella spiralis*. The eradication of *Echinococcus granulosus*, which is conveyed in the offals to dogs, who in turn transmit it to man, would not, however, be greatly assisted by this method and the parasite will remain a danger to dog owners so long as the feeding of uncooked slaughterhouse offals to dogs is countenanced and sheep dogs have the opportunity to feed on the carcasses of sheep that have died in the less accessible parts of marginal or hill land.

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# Packaging for Irradiated Foods

G. E. TRIPP

Food Radiation Preservation Division, Quartermaster Food and Container Institute for the Armed Forces, Chicago, Illinois

Packages for irradiated foods must be compatible with the radiation treatment, the food and the production line. In the case of foods treated by radiation doses of 1 Mrad, or less, common commercial packages appear satisfactory. Radiation doses of more than 1 Mrad have an adverse effect on packages and care must be taken in selecting packages for foods to be treated by radiation doses in excess of 1 Mrad.

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THE purpose of this paper is to describe the important effects of radiation on packaging and packing materials. The factors that are of major concern to the research worker or technologist involved in packaging problems connected with the radiation preservation of food are:

(1) the adaptability of the container, whether rigid or flexible, to the source of radiation treatment and, as a natural corollary, the capacity of the container to preserve the irradiated food during handling and storage,
(2) the capacity of the container material to accept, without undue injury to its chemical composition and physical structure, periods of exposure to $\gamma$-rays or high energy electrons,
(3) the imperviousness of the container, its components (such as seals) and the container material to the entry of spoilage organisms,
(4) the capacity of the container material to submit to radiation without imparting off-odors or flavors to the contents of the food package, and
(5) miscellaneous properties that in one degree or another are associated with the four factors listed.

In this review of progress on the packaging aspect of radiation preservation of foods no attempt will be made to give in detail the substance of the many articles and comprehensive treatises on the effects of radiation on materials. For our purposes here the status of packaging for irradiated foods can best be defined under the foregoing headings.

## PACKING WITH RESPECT TO THE "RADIATION PRODUCTION LINE"

The first topic, therefore, is the adaptability of the container to a "radiation production line" and its capacity for preserving food during handling and storage. This is an appropriate place to begin because we ought to visualize at the outset what manner of package we might be dealing with.

Within the limitations discussed below tin-plate cans, plastic coated aluminum containers and plastic containers appear satisfactory for irradiated foods. The shape of the container will be dictated by the configuration of the radiation source; the maximum dimensions of the container will be determined by the size of the source, energy of the radiation and desired dose distribution within the package. Ideally, it would be desirable to have a uniform distribution of dose within the package ensuring that all of the contents receive an identical dosage. This is not possible because of the absorption of radiation in matter and because of the emission of secondary radiation in the form of X-rays or Compton electrons. At present

a dose distribution of 100 per cent to 125 per cent is acceptable. This requirement partially dictates the thickness of our package.

*Electron source*

In the case of irradiation with high energy electrons, the electrons have a limited range. The maximum range of electrons of a given energy can be calculated on the basis of the following equation

$$R_{max} = \frac{0{\cdot}542E - 0{\cdot}133}{d}$$

$R_{max}$ is the maximum range in cm
$E$ is the energy of electrons in MeV
$d$ is the density of irradiated material in g/cm².

The above equation cannot be used directly since it describes a distribution of dose from zero to 100 per cent. We must use, therefore, a value known as the "effective range" which is equal to 3/5 $R_{max}$. Using this value we find that the thickness of the package will be 0·33 g/cm²/MeV if the irradiation is conducted from one side. If we bombard the package from two sides the thickness will be 0·81 g/cm²/MeV.

The electron accelerators which have been used for food irradiation in the United States employ a beam of electrons which is manipulated to scan the package. The width of the package is determined by the limits of the scanning beam, the thickness by the absorber density of the contents and the energy of the electrons and the length by convenience or weight limitations. These limitations describe a rectangular package. Cylindrical containers could be irradiated in devices constructed in this manner; however, 20 per cent of the radiation would be wasted and production decreased by a like amount.

*Gamma source*

The bulk of the irradiated foods produced under the Quartermaster radiation program have been irradiated in $\gamma$-sources consisting of a circular array of spent fuel elements.

This source configuration is ideally suited to a cylindrical container. Theoretical considerations reveal that the dose distribution of $\gamma$-rays in cylindrical cans, exposed in a cylindrical source of infinite length, would conform to the following equation:

$$D = D_0(1 + 1/10(r/R))^2$$

$D_0$ is the dose at the center of the can
$D$ is the dose at some point along the equatorial plane
$r$ is the radius of a point along the equatorial plane
$R$ is the can radius.

Dose measurements indicate that experimental values are in close agreement with theoretical results. Considerations are being given to the use of flat, parallel $Co^{60}$ plates as a $\gamma$-source for the United States Army Ionizing Radiation Center. Considerations of the geometry indicate that rectangular packages would be most desirable.

Having determined the shape of the container we must now consider the capabilities of the container to preserve the irradiated food during handling and storage.

## PRESERVATION CAPABILITIES

Numerous studies have been made with tin-plate containers. One such study involved red sour pitted cherries, chile, green beans, beef, pork and fish packed in cans constructed with various tin coating weights and interior enamels. Less thorough studies have been conducted with peas, spinach, bacon, chicken, shrimp, carrots, peaches and maize. These studies demonstrated that the commercial steel and tin coating weight used for commercial thermal processed foods would also be suitable for their irradiated counterpart. Further, it was noted that precautions must be taken in the selection of end sealing compounds and, for a few foods, with the interior enamels. These precautions will be discussed later.

Generally, with the exceptions noted, little difference has been observed in the effects of irradiated or thermal processed food on container shelf life. We can, therefore, expect that most cans suitable for

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thermal processed foods will provide a similar shelf life to its irradiated counterpart.

*Tin-plate cans*

The ability of tin-plate cans to resist rough handling is well known and these attributes are not affected by radiation. Where irradiation is performed under water precautions must be taken to avoid a high internal vacuum in the can. Too large a pressure differential can result in collapsing of the can.

*Flexible containers*

During storage tests of irradiated foods in flexible containers it was observed that foods can have an adverse effect on the package, leading to package failure (Table 1). The failures varied, in some cases appearing as delamination of laminated materials, package leakage and in some cases splitting adjacent to the sealed areas. Subsequent investigations revealed that the leakage and splitting were caused by stress-cracking promoted by fats and free fatty acids. In part the failures that were observed have been caused by the time delay encountered between packaging and irradiation treatment. This delay occurred because the radiation facility was located a long distance from the research group who prepared the samples. Since irradiation improves the stress-cracking resistance of plastics, such as polyethylene, we can expect fewer losses where the irradiation is performed shortly after packaging.

As part of the program of evaluating materials, one flexible package has been found, which, except for odor, appears satisfactory for irradiated foods. The package consists of a polyethylene coated polyester film. During tests in which this material was in contact with cottonseed oil over a six-month period, it did not delaminate or show signs of failure. Further, when this material was tested with 5% lactic acid solution, stress-cracking did not occur. Currently this polyethylene–polyester combination is being laminated to aluminum foil and tested as a potential flexible package for irradiated foods.

The polyethylene–polyester lamination has been subjected to laboratory rough handling tests to determine if it will provide protection during shipment. The tests have been conducted using mock-up packages containing water, packed in shipping cases. Artificially produced rough handling by drum and drop test showed that the container, when packed in properly designed shipping containers, has surprising durability (Table 2). Results of these tests indicate that this type package has sufficient durability for overseas shipment. The tests were carried to the point of failure. The failures occurred at points coinciding with multiple folds and creases.

TABLE 1. Failures of flexible packages (%)

| Package | Contents | Dose (Mrads) | | | |
|---|---|---|---|---|---|
| | | 0 | 1 | 3 | 6 |
| Polyethylene (H.D.) | Lactic acid sol. | 40 | — | — | — |
| | Salad oil | 25 | — | — | — |
| Nylon (caprolactam) | Lactic acid sol. | 80 | 80 | 83 | 83 |
| Polyvinyl chloride | Salad oil | 20 | 20 | 20 | 20 |
| -Fluorothane | Salad oil | 20 | 20 | 50 | 100 |
| Polyethylene laminated to cellophane | Lactic acid sol. | 0 | 33 | 20 | — |
| | Salad oil | 100 | — | — | — |
| Polyethylene coated Mylar | Lactic acid sol. | 25 | 35 | 50 | 35 |
| Saran 6D | Lactic acid | 80 | 95–100 | 95–100 | 95–100 |

The occurrence of folds and creases and the susceptibility of this portion of the package to cumulative damage is of concern because penetration by micro-organisms may occur.

TABLE 2. Revolving drum tests of flexible containers packed in shipping containers

| Package material | | Falls to failure (leakage) | |
|---|---|---|---|
| | | Minimum | Maximum |
| Polyethylene coated Polyester film type "S" | Test 1 | 340 | 450 |
| | Test 2 | 509 | 600+ |
| Polyester, type V, (0·0005 in.) Laminated to aluminum (0·001 in.) Laminated to polyester, type M (0·0005 in.) | | 230 | 270 |

## EFFECT OF RADIATION ON MATERIALS

We now turn our attention to the second factor: the capacity of the package to accept without undue injury to its chemical composition and physical structure, periods of exposure to $\gamma$-rays or high energy electrons.

When plastics and other organic materials are exposed to high energy radiation, ions and free radicals form in the solid material. This results in cross-linking or chain scission. Simultaneously, unsaturated bonds, hydrogen and other gases may be formed, and where the irradiation occurs in air, oxidation may also occur. The degree of change observed depends upon the radiation dose and the chemical nature of the polymer. Hydrocarbon polymers are least affected by radiation, while the halogenated plastics and cellulosics are degraded.

Organic substances are used in the manufacture of tin-plate cans. They comprise the small amount of rubbery substance applied to the end of the can, known as end sealing compound, and interior enamels. End sealing compounds formulated with butyl rubber should be avoided because of degradation during irradiation which could lead to can failure. In the case of interior can enamels, indications are that the enamels are improved by radiation. However, enamels containing zinc pigments should not be used for peas and green beans since these irradiated items will leach the zinc pigment from the enamel. This difficulty can be avoided by omitting the pigment.

The radiation dose used in sterilizing food (4·8 Mrads) produces slight changes in the physical properties of the plastic used in fabricating the package. The changes are small and will not preclude the use of plastics from the standpoint of strength considerations. For example, cellulosics, which are degraded by radiation, show a decrease of only 10 per cent in tensile and bursting strengths.

## IMPERVIOUSNESS

The third factor, that of the imperviousness of the container, its components such as seals, and the container material to the entry of spoilage organisms, is a major factor, particularly in the case of flexible containers.

*Tin-plate cans*

Tin-plate cans have been in use for nearly 100 years and have an admirable record in preserving foods from spoilage organisms. The only precaution necessary, in respect to

irradiated foods is the avoidance of compounds formulated with butyl rubbers.

*Flexible containers*

During the manufacture of flexible containers, defects occur which can compromise the package as a barrier to micro-organisms. Single plastic films 1/1000 in. thick or less are readily penetrated by micro-organisms. Films 1/1000 to 2/1000 in. thick are not penetrated. However, creasing damages the film permitting entry of micro-organisms. Packages made of single films thicker than 2/1000 in. are resistant to penetration, even after creasing. It has also been shown that laminated films are more susceptible to damage than single ply films of the same thickness. In these tests, the creases were prepared by a standard creasing technique which is quite severe but did not involve repetitive damage. Studies using a less severe method of creasing and involving repetitive damage are now in progress. Preliminary data indicate that our current testing procedure correlates with the larger scale rough handling tests (Table 3).

TABLE 3. Dye penetration after flexing (Gelbo tester)

| Package | Number of flexes before failure |
|---|---|
| Saran (0·002 in. thick) | 300 |
| Polyethylene (low density 0·002 in. thick) | 500+ |
| Polyethylene (0·002 in.) coated polyester (0·0005) type S | 200 |
| Polyethylene (0·002 in.) coated polyester (0·001 in.) type M | 500+ |
| Polyvinyl chloride (0·0015 in.) laminated to aluminum foil (0·001 in.) laminated to polyester (0·0005 in.) type M | 50 |

In this connection it should be noted that radiation does not play a part in these failures. The failures are an inherent property of the material.

## OFF-FLAVOR OFF-ODOR CONSIDERATIONS

TABLE 4. Gaseous products ($\mu$moles/g sample) (dose 6 Mrads)

| | Polyethylene (low density) | | Polyethylene (high density) | | Fluorothane | |
|---|---|---|---|---|---|---|
| | Vacuum | Air | Vacuum | Air | Vacuum | Air |
| Carbon dioxide | 0·3 | 0·3 | 0·1 | 0·1 | 27·9 | 27·7 |
| Oxygen | 0·1 | 0·1 | 0·1 | 0·1 | 0·3 | 0·4 |
| Nitrogen | 1·5 | 1·5 | 0·8 | 0·8 | 8·5 | 8·1 |
| Water | 0·5 | 3·8 | 0·2 | 1·1 | 0·5 | 2·1 |
| Hydrogen | 95·7 | 93·6 | 98·7 | 94·6 | 0·6 | — |
| Silicon tetrafluoride | — | — | — | — | 59·9 | 57·5 |
| Hydrogen chloride | — | — | — | — | 2·3 | — |
| Ethylene | 1·2 | — | — | — | — | — |
| Acetone | 0·3 | — | — | 3·2 | — | 4·3 |
| Butene | 0·1 | 0·6 | — | — | — | — |
| Propene | 0·3 | — | 0·1 | — | — | — |

TABLE 5. Gaseous products ($\mu$moles/g sample)
(not corrected for residual air)
(dose 6 Mrads)

| | Saran | | Mylar | |
|---|---|---|---|---|
| | Vacuum | Air | Vacuum | Air |
| Carbon dioxide | 6·2 | 1·3 | 0·1 | 0·5 |
| Argon | — | 1·1 | 0·9 | 1·0 |
| Oxygen | — | 4·8 | 20·3 | 18·6 |
| Nitrogen | 5·5 | 92·0 | 78·4 | 79·4 |
| Water | 5·1 | 0·4 | 0·3 | 0·3 |
| Hydrogen | 5·2 | 0·1 | — | 0·2 |
| Silicon tetrafluoride | 1·1 | — | — | — |
| Acetic acid | 12·2 | — | — | — |
| Ethanol | 0·6 | — | — | — |
| Hydrogen chloride | 43·5 | 0·3 | — | — |
| Hydrogen cyanide | 18·4 | — | — | — |
| Ethylene | 2·2 | — | — | — |

TABLE 6. Odor intensity of irradiated plastics

| Plastic | Dose (Mrads) | | | | |
|---|---|---|---|---|---|
| | 0 | 1 | 3 | 6 | Average |
| Polyethylene (no additives) | 1·6 | 3·2 | 3·6 | 4·7 | 3·3 |
| Polyethylene (commercial) | 1·4 | 4·0 | 5·2 | 5·6 | 4·1 |
| Polyethylene (2·5 per cent carbon black plus anti-oxidant) | 1·6 | 4·2 | 5·5 | 6·0 | 4·3 |
| Polyethylene (6·0 per cent carbon black plus anti-oxidant) | 2·6 | 4·4 | 5·2 | 5·4 | 4·4 |
| Polyethylene (H.D. Ziegler process) | 1·7 | 3·8 | 3·6 | 3·9 | 3·2 |
| Wax (Fischer Tropsch) | 2·0 | 3·7 | 4·6 | 5·3 | 3·9 |
| Polystyrene | 3·0 | 2·4 | 2·8 | 1·6 | 2·5 |
| Nylon 6 | 1·9 | 1·4 | 1·7 | 1·4 | 1·6 |
| Mylar | 2·2 | 2·0 | 2·6 | 2·5 | 2·3 |
| Videne | 1·6 | 2·5 | 2·4 | 2·1 | 2·2 |

Factor four, the capacity of the container material to submit to radiation without imparting off-flavors or odors to the contents of the food package, describes another area where flexible containers are deficient.

Gaseous products are formed in plastics during exposure to radiation. The gases consist of hydrogen, carbon dioxide, water, carbon monoxide and other organic and inorganic gases (Tables 4 and 5). These gases are produced in quantities large enough to be detected in the contents of plastic bags. For example, chloride ions to the extent of 120 p.p.m. have been found in water contained in polyvinylchloride bags exposed to 6 Mrad $\gamma$-radiation. The organic gases result in objectionable odors which can be acquired by the food. Of the plastics used in packaging polystyrene develops the least odor; the polymides (nylon,

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Rilsan) and polyesters (Videne, Mylar) are also low in odor. Polyethylene is particularly objectionable (Table 6). In the case of polyethylenes the aliphatic side chain appears to be responsible since the odors observed after irradiation are more intense with the highly branched low density polyethylene than with the linear, high density variety. Less volatile materials are produced in addition to the gaseous products. These are extracted by the contents of the package and contribute to off-flavors (Table 7).

TABLE 7. Non-volatile water soluble substances in irradiated polyethylene (H.D.) ($\mu$g/g of film)

| Dose (Mrads) | | | |
|---|---|---|---|
| 0 | 1 | 3 | 6 |
| 27·4 | 29·8 | 31·2 | 37·1 |

## MISCELLANEOUS PROPERTIES

In respect to factor five, miscellaneous properties that in one degree or another are associated with the four factors listed above, unusual conditions arising from the process have an effect on the container.

It is possible to preserve fresh meats with this process. However, the enzymes contained in raw meats are not destroyed. The fat splitting enzymes produce free fatty acids which attack interior can enamels compounded with oleoresinous enamels. This can be avoided by use of phenolic or epoxy enamels or by inactivating the enzymes.

Other possible problems exist in this area. One is the possibility that the extractives from can enamels and plastic films may be toxic. This is quite remote since animal feeding studies have included the use of enamel lined cans and plastic bags and to date no evidence of container toxicity has been observed. A second possibility is that of induced radioactivity of tin plate containers during exposure to high energy electrons (16–24 MeV). This latter is currently under study.

## PASTEURIZING DOSES

The previous discussion has been directed to food containers for use with food "sterilized" by radiation (4·8 Mrad dose for most foods). Only very limited packaging studies have been undertaken with foods "pasteurized" with ionizing radiation of 1 Mrad or less. According to the work that has been conducted with packages at higher doses, the effect of ionizing radiation at 1 Mrad or less appears negligible. The selection of the package, in this case, will depend upon the objective of the radiation treatment, anticipated shelf life, end use and container cost.

*Present capabilities*

The commercial tin-plate can is readily adaptable to the radiation process and has a long history of rugged satisfactory performance with foods. The tin-plate can could be used for irradiated foods with a high degree of confidence. Flexible containers demonstrate a good potential as packages for irradiated foods. However, this is a new application for flexible containers, therefore a considerable amount of experimentation and study is required before we can have the same degree of confidence in flexible containers.

Current considerations for the source configurations of the U. S. Army Ionizing Radiation Center are suited to rectangular shaped containers. Rectangular tin-plate cans and flexible containers are available.

*Effect on foods*

Flexible containers release materials capable of imparting off-flavors and odors to foods. Current studies indicate that this can be controlled.

*"Pasteurized" food*

Packaging does not present a problem for foods "pasteurized" by radiation. Current commercial packages should prove adequate.

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# The Economics of Machine Sources of Radiation

M. C. CROWLEY-MILLING
Metropolitan-Vickers Electrical Co. Ltd., Trafford Park, Manchester 17

The correlation between size and cost of existing electron accelerators is examined, and the total running cost of a k5W linear accelerator installation is shown to be about 14s. to 27s. per kWh. The possible efficiency of utilization is discussed and a table gives expected costs for a number of applications. A sketch shows an installation suitable for treating boxes on a conveyor system.

THE economics of machine sources depend on the cost of radiation power and the efficiency of utilization.

The capital cost of existing electron generators has been plotted against power output, and is shown in Fig. 1. The cost per kW is seen to be approximately £33,000/(kW)$^{0.6}$ except for low energy machines, which are somewhat cheaper.

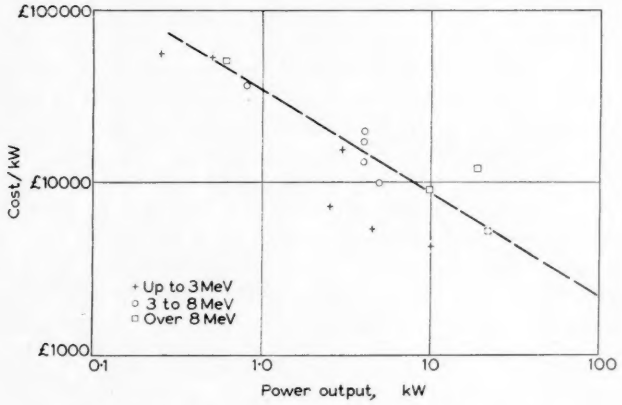


FIG. 1. Variation of cost/kW with size of machine.

Taking a 6 MeV, 5 kW linear accelerator as typical of the type of machine which would be used for large scale food irradiation, an estimate of running costs has been made and this is given in 1(a) Tables and 1(b). This estimate includes depreciation and interest on the capital cost of the equipment, the building and conveyor, and the provision of spares. It can be seen that the running cost would be about 27 s. per kWh for single shift working, and about half this for continuous operation. This is equivalent to 0·4 penny and 0·2 penny per Mrad-lb if fully utilized. Other published estimates of the cost of electron power give figures varying between 0·1 and 1·0 penny per Mrad-lb, depending on the size of machine and the method of accounting.

The efficiency of utilization depends on the area and depth efficiencies. For uniform materials, the depth efficiency for irradiation

TABLE 1(a). Running costs of electron accelerator

| Capital cost | £ | Hourly running costs | £ | s. | d. |
|---|---|---|---|---|---|
| 6 MeV, 5 kW linear accelerator | 50,000 | Klystron replacement | | 15 | 0 |
| Building and conveyor | 20,000 | Modulator valves | | 10 | 0 |
| Spares | 4,000 | Other valves and components | | 5 | 0 |
| | | Skilled maintenance (part time) | | 3 | 0 |
| | | Miscellaneous | | 3 | 6 |
| | | Electric power 42 kW @ 1d | | 3 | 6 |
| | 74,000 | | 2 | 0 | 0 |

from one side only is a maximum, about 70 per cent, when the electron energy in MeV is approximately equal to:

$$7{\cdot}5 \times (\text{thickness of material in in.}) \times (\text{specific gravity of material})$$

over the energy range 2–20 MeV. If irradiation can be carried out from both sides, material about 2·5 times as thick can be treated, and the depth efficiency can reach 90 per cent. Area efficiency depends on the shape and uniformity of the material, and can reach 90 per cent for bulk material in boxes, on a conveyor system, or passing through tubes. Thus overall efficiencies up to 60 per cent for single side irradiation and 80 per cent for irradiation from both sides can be reached, although for many applications the efficiency will be appreciably below these figures.

Taking this into consideration, Table 2 shows the approximate cost of irradiation for a number of different processes, assuming operation of the equipment for 40 hr per week only. The costs would be halved for continuous use.

A suitable layout for the irradiation of uniform rectangular boxes of foodstuffs is shown in an artist's impression in Fig. 2. Here it is assumed that an irradiation unit is to be inserted in an existing conveyor system with as little disturbance as possible, and that the thickness of the boxes is such that irradiation from both sides is advisable. The irradiation chamber is underground, to ease the problem of radiation protection, and the linear accelerator is mounted above the chamber, its electron beam directed downwards through a spreading device, which spreads the beam to cover the full width of the conveyor. The boxes are carried down to the irradiation chamber by a sloping conveyor and transferred to the positive drive conveyor which carries them through half the beam, turns them over at the end, and carries them back through the beam to expose the other side. The boxes are then carried back to the main conveyor system. Other conveyor arrangements could be made for different packaging of the food, or for bulk material, and it is possible to split the beam and irradiate from both sides simultaneously for cases, such as liquids, where inversion of the package does not

TABLE 1(b). Total costs

| | 40 hr per week | Continuous service |
|---|---|---|
| Depreciation on capital (10 yr basis) | £ 3 14 0 per hr | £ 18 3 per hr |
| Interest on capital (5% on £37,000) | 18 6 per hr | 4 6 per hr |
| Running costs | 2 0 0 per hr | 2 0 0 per hr |
| | 6 12 6 per hr | 3 2 9 per hr |
| Equivalent to about: | 0·4d per Mrad-lb. | 0·2d per Mrad-lb. |

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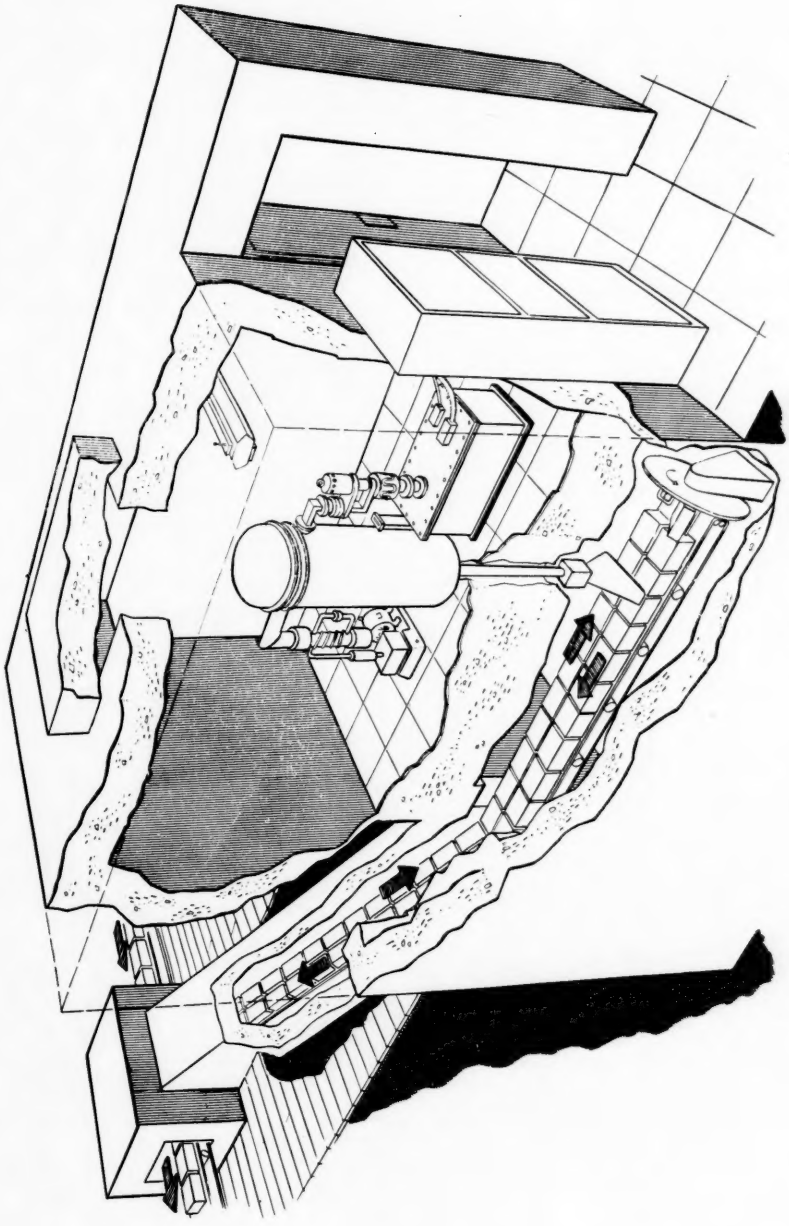

FIG. 2.

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TABLE 2

| Process | Typical dose (Mrad) | Estimated utilization (%) | Capacity of 5 kW machine | Approx. cost* of treatment |
|---|---|---|---|---|
| Prevention of potato sprouting | 0·01 | 50 | 90 tons/hr | 1/6d/ton |
| Disinfestation of grain | 0·05 | 80 | 28 tons/hr | 4/8d/ton |
| Improving the keeping power of foodstuffs | 0·05 | 40 | 14 tons/hr | 9/4d/ton |
| Sterilization of foodstuffs | 0·5<br>2·0 | 40<br>40 | 3200 lb/hr<br>800 lb/hr | ½d/lb<br>2d/lb |

* Machine running 40 hr per week: continuous running would halve the cost.

necessarily cause inversion of the contents.

The widespread application of irradiation for food preservation depends on the development of methods of preventing unwanted side effects, and not on the economics of irradiation power. As can be seen from Table 2, at present day costs some of the processes are economically attractive, and it is expected that the next few years will see an appreciable reduction in the cost of electron power, through simplification of the apparatus and longer life of components.

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# Economics of Gamma Radiation Processing

G. S. MURRAY
Technological Irradiation Group, Isotope Division, Wantage Radiation Laboratory, Wantage, Berks

The availability, cost and utilization of $\gamma$-radiation sources are discussed. The effects on radiation processing costs of variations in these factors and in the scale of operation are considered. It is concluded that probable costs do not preclude immediate applications if plant is efficiently designed and processing is on a sufficiently large scale.

## INTRODUCTION

THE technical feasibility of some radiation processes is well on the way to being established and while it is unlikely that food treatments, other than at very low doses, will be acceptable for a few years, there are impending applications in sterilization of surgical, medical and pharmaceutical products and in the disinfecting of animal fibres and other materials.

In the general field of radiation processing the type of plant required will vary with the application, but in the particular fields mentioned there is an immediate requirement for package irradiation equipment. During the past year at Wantage Radiation Laboratory the design of such equipment has been studied and developed, and a pilot plant capable of handling 7 Mrad–tons per day is now in construction. The plant will handle packages of about 1 $ft^3$ in volume at various rates of throughput so that total radiation doses from 10,000 rads to 5 Mrads will be available.

Concurrent with the development of this plant there have been many discussions with potential users of package irradiation processes and it is hoped that the familiarity thus gained with the problem will make possible a realistic assessment of economics.

## SOURCES OF $\gamma$-RADIATION

Radioisotopes emitting $\gamma$-rays are produced in a nuclear reactor either as by-products of the fission reaction or by neutron capture in other isotopes. There have been many speculative estimates of radiation processing costs associated with the use of spent fuel elements, fission product gases, reactor loops and the like, but at Wantage we have been particularly concerned with the possible use of $Cs^{137}$ and $Co^{60}$.

By early 1961 the U.K.A.E.A. intends to have in operation a plant which will separate rather more than 1 Mc of the fission product $Cs^{137}$ each year. The nuclear power programme could also provide Mc quantities of $Co^{60}$ as a by-product of normal reactor operation. The Wantage pilot will use about 150,000 c of $Co^{60}$ and it is also intended to ship a similar quantity in 1959 to Australia where it will be used in an industrial process. This paper will therefore concentrate on attempting an estimate of costs based on the use of these two isotopes which offer immediate prospects of application.

Caesium-137 decays with a half-life of about 27 years and to maintain a source at its original strength requires 10 per cent make up every 4 years. Eighty-two per cent of disintegrations result in the emission of a $\gamma$-ray of 0·662 MeV so that the mean energy available per disintegration is 0·54 MeV. One megacurie of $Cs^{137}$ will provide 3·2 kW of $\gamma$-energy.

Cobalt-60 decays with a half-life of 5·2 years and to maintain a source at its original strength will require 12·5 per cent make up per annum. Each disintegration results in the emission of 2 $\gamma$-rays with energies of 1·17 and 1·33 MeV. One megacurie of $Co^{60}$ will provide 14·8 kW of $\gamma$-energy.

Because the $\gamma$-rays from caesium have less energy than those of cobalt there will in general be more self-absorption in the source and to a good approximation the effective emission of energy per c of cobalt will be at five times the rate per c of caesium.

*Efficiency in the use of source radiation*

The source strength which is required for a specific throughput and treatment depends not only on the intensity of radiation but also on the proportion of energy which can be effectively absorbed in the process material. The higher this proportion the lower will be the source strength and cost.

Because $\gamma$-radiation is emitted isotropically it is important to use as much as possible of the whole solid angle round the source.

A proportion of the radiation incident on the product will pass through and be wasted. To keep this loss to a minimum the depth of product treated should be as large as is practicable. Also the higher the density of the product the more energy it will absorb for a given depth or thickness.

Another factor is the variation of energy deposition or dose through the product. The intensity of radiation falls off with distance from the source and is further reduced by absorption so that even by irradiating from both sides there will be a variation in dose throughout the process material. A specific treatment will require a minimum dose. A maximum dose may also be specified but energy deposited above the minimum requirement is effectively wasted.

In general as the thickness of product presented to the source decreases, the less will be the variation in dose. The variation also decreases with distance from the source.

Some of the factors affecting efficiency militate against others and the best compromise is achieved by careful design of the geometrical relationship of source and process material.

Experimental work on a model system at Wantage Radiation Laboratory, using a $Co^{60}$ source of variable dimensions, has shown that it is practicable to realize an efficiency of 40 per cent with standard packages of about 1 $ft^3$ in volume and density 0·5.

Higher efficiencies can be achieved by decreasing package size or increasing density and the converse also applies. If the packages vary in size this may have a detrimental effect on efficiency. Theoretical estimates are in fair agreement with these experiments and indicate that similar efficiencies may be achieved with $Cs^{137}$.

*Size of source*

The size of source will depend on the total dose required, the rate of throughput and the efficiency. The following total doses will be considered:

(1) 20 krads corresponding to disinfestation or sprouting inhibition.
(2) 200 krads corresponding to pasteurization.
(3) 2 Mrads corresponding to sterilization or disinfection.

Four rates of throughput will be assumed: $2{\cdot}5 \times 10^2$, $2{\cdot}5 \times 10^3$, $2{\cdot}5 \times 10^4$, $2{\cdot}5 \times 10^5$ lb/hr. Efficiency will be taken as 40 per cent.

The number of curies of $Cs^{137}$ required for a specific dose and throughput will be five times the number of curies of $Co^{60}$. Cobalt sources corresponding to various doses and throughputs are tabulated below.

TABLE 1. Size of source (c of $Co^{60}$)

| Throughput lb/hr | Dose | | |
|---|---|---|---|
| | $2 \times 10^6$ rads | $2 \times 10^5$ rads | $2 \times 10^4$ rads |
| $2{\cdot}5 \times 10^2$ | $10^5$ | — | — |
| $2{\cdot}5 \times 10^3$ | $10^6$ | $10^5$ | — |
| $2{\cdot}5 \times 10^4$ | $10^7$ | $10^6$ | $10^5$ |
| $2{\cdot}5 \times 10^5$ | — | $10^7$ | $10^6$ |

*Source costs*

The cost of separating $Cs^{137}$ will depend on the scale of operation. No figures are yet available for the actual large scale production but various estimates have been made. In America, costs including handling and shipping have been estimated from 10 cents to several dollars per c. In the United Kingdom costs arising from the planned separation plant are unlikely to exceed 10s. per c. Costs ranging from 10s. to 2s. 6d. per c will therefore be considered; 10s. representing the upper limit when large scale production begins and 2s. 6d. the probable order of costs in about 10 years.

The cost per c of $Co^{60}$ will depend to some extent on the specific activity (c/g), but in general is likely to be similar to the cost of caesium. Figures between \$1.00 and \$2.00 per c have been frequently quoted and in the United Kingdom quantities in excess of 100,000 c are expected to be in the same range of prices in the immediate future. The regular production of $Co^{60}$, on a large scale from excess neutrons in nuclear power programme reactors, may result in lower costs and it is therefore proposed to consider the same range from 10s. to 2s. 6d. per c as for $Cs^{137}$.

In estimating the annual cost of a radiation source it must be remembered that the source can only be used effectively for part of its life. It will be assumed that after two half-lives the specific activity is not acceptable and thus only 75 per cent of the original energy can be used.

Cobalt-60 decays by 12·5 per cent per annum which is equivalent to about 17 per cent of the available energy. Adding interest at 6 per cent the total annual source cost may be taken as 23 per cent of capital value.

Similarly $Cs^{137}$ decays by 2·5 per cent per annum which is equivalent to $3\frac{1}{3}$ per cent of the available energy. Adding interest at 6 per cent the total annual source cost is $9\frac{1}{3}$ per cent of capital value.

At a fixed efficiency the source cost per lb of product will be independent of the rate of throughput and depend only on the total dose required. Tabulated below are costs in pence/lb for various total doses arising from costs per c of 10s., 5s. and 2s. 6d.

*Other costs*

Other costs arise from depreciation and interest on plant, labour, overheads, maintenance and power.

The plant consists of the following components,

(1) A radiation room enclosed by shielding
(2) Mechanical handling equipment for conveying the process material into the radiation room, round the source and out again
(3) A shielded store in which the source can be placed when access to the radiation room is required
(4) An interlock system to prevent access to the radiation room except when the source is safe
(5) A control system
(6) Sundry services including provision for loading and maintenance of the source.

The labour required will be not less than four operators, one on each of three 8 hr shifts per day, plus one supervisor. At a throughput of 250 lb/hr the operator will load process material manually on to the

TABLE 2. Source costs (pence/lb)

| Total dose (rads) | $Co^{60}$ cost/c | | | $Cs^{137}$ cost/c | | |
|---|---|---|---|---|---|---|
| | 10s. | 5s. | 2s. 6d. | 10s. | 5s. | 2s. 6d. |
| $2 \times 10^6$ | 1·4d | 0·7 | 0·35 | 2·8 | 0·4 | 0·7 |
| $2 \times 10^5$ | 0·14 | 0·07 | 0·035 | 0·28 | 0·14 | 0·07 |
| $2 \times 10^4$ | 0·014 | 0·007 | 0·0035 | 0·028 | 0·014 | 0·007 |

TABLE 3. Partial, source ($Co^{60}$) and total costs (pence/lb)

| Throughput lb/hr | Total dose | | | | | | | |
|---|---|---|---|---|---|---|---|---|
| | $2 \times 10^6$ rads | | | $2 \times 10^5$ rads | | | $2 \times 10^4$ rads | |
| | $2{\cdot}5 \times 10^2$ | $2{\cdot}5 \times 10^3$ | $2{\cdot}5 \times 10^4$ | $2{\cdot}5 \times 10^3$ | $2{\cdot}5 \times 10^4$ | $2{\cdot}5 \times 10^5$ | $2{\cdot}5 \times 10^4$ | $2{\cdot}5 \times 10^5$ |
| Source cost (10s./c) | 1·4 | 1·4 | 1·4 | 0·14 | 0·14 | 0·14 | 0·014 | 0·014 |
| Partial cost | 1·6 | 0·3 | 0·06 | 0·24 | 0·05 | 0·02 | 0·04 | 0·015 |
| Total cost | 3·0 | 1·7 | 1·46 | 0·38 | 0·19 | 0·16 | 0·054 | 0·029 |
| Source cost (5s./c) | 0·7 | 0·7 | 0·7 | 0·07 | 0·07 | 0·07 | 0·007 | 0·007 |
| Partial cost | 1·6 | 0·3 | 0·06 | 0·24 | 0·05 | 0·02 | 0·04 | 0·015 |
| Total cost | 2·3 | 1·0 | 0·76 | 0·31 | 0·12 | 0·09 | 0·047 | 0·022 |
| Source cost (2s. 6d/c) | 0·35 | 0·35 | 0·35 | 0·035 | 0·035 | 0·035 | 0·0035 | 0·0035 |
| Partial cost | 1·6 | 0·3 | 0·06 | 0·24 | 0·05 | 0·02 | 0·04 | 0·015 |
| Total cost | 1·95 | 0·65 | 0·41 | 0·275 | 0·085 | 0·055 | 0·0435 | 0·0185 |

input conveyor in addition to controlling the process. At higher throughputs process material will be automatically loaded from an input store, and conveyed to an output store after treatment.

The plant will operate 160 hr per week and 50 weeks per yr. This will allow one day shift per week for maintenance and 2 weeks per yr for annual overhaul and source make up.

The size and cost of the plant will be determined largely by the dimensions of the source and to some extent by the rate of throughput. Three typical examples of partial annual operating costs are estimated below.

(i) 250 lb/hr at 2 Mrads

| | | |
|---|---|---|
| Plant capital cost | | |
| Civil engineering | 25,000 | |
| Mechanical and electrical engineering | 25,000 | |
| Total | £50,000 | |
| Partial annual operating cost | | |
| depreciation @ 10 per cent | 5,000 | |
| interest average 3 per cent | 1,500 | 6,500 |
| Labour | | |
| 4 operators at £600 | 2,400 | |
| 1 supervisor (half-time) | 600 | 3,000 |
| overhead | | 2,000 |
| maintenance | | 1,000 |
| power | | 500 |
| Total | | £13,000 |

250 lb/hr, 8,000 hr per year is equivalent to $2 \times 10^6$ lb/annum cost per lb = 1·6 pence.

(ii) 2,500 lb/hr at 200 krads

Source size and plant size as in (i) with the addition of input and output storage and automatic loading.

| | | |
|---|---|---|
| Plant capital cost | £70,000 | |
| Annual operating cost | | |
| depreciation and interest | | 9,100 |
| Labour | | |
| 4 operators @ £600 | 2,400 | |
| 1 supervisor | 1,200 | 3,600 |
| overhead | | 2,400 |
| maintenance | | 2,400 |
| power | | 2,500 |
| Total | | £20,000 |

cost per lb = 0·24 pence.

(iii) 2500 lb/hr at 2 Mrads
Source size and therefore plant size increased.
Plant capital cost £100,000
Maintenance and power slightly increased partial annual operating cost £25,000
cost per lb = 0·3 pence.

To such partial costs per lb must be added source cost per lb and this information is summarized in Table 3. Source costs for $Co^{60}$ only are shown. Source costs for $Cs^{137}$ are at twice the rate for the same cost per c.

## CONCLUSIONS AND GENERAL OBSERVATIONS

(1) All the above costs are feasible in relation to some processes and offer a reasonable prospect of immediate application.

(2) Source costs are a significant proportion of total costs and emphasize the need for careful design to attain a high efficiency particularly at high total doses.

(3) Partial costs (i.e. other than source costs) fall rapidly as the rate of throughput increases.

(4) Rates of throughput below 250 lb/hr are unlikely to be economic except for very high costs products.

The above estimates of costs are all related to package irradiation equipment. Where possible, the bulk handling of high rates of throughput as for example in disinfestation of grain, could lead to substantial reductions in cost.

The $\gamma$-rays from $Co^{60}$ and $Cs^{137}$ do not induce radioactivity in any process material.

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# Économie Comparée des Procédes de Conservation des Aliments Utilisant le Froid et la Chaleur

M. ANQUEZ* et A. BÉNÉZIT†
Section Technique du Froid, Ministère de l'Agriculture,
19 Avenue du Maine, Paris (15e) France

Les auteurs donnent d'abord quelques indications relatives au coût de l'entreposage frigorifique en France et dans différents pays d'Europe.

Ils comparent ensuite les prix de revient de la conservation de viandes et de petits pois congelés et appertisés (stérilisés à la chaleur). Ces prix de revient sont décomposés en plusieurs postes: fabrication, stockage, transport et distribution.

Dans le cas de la viande, la comparaison doit être interprétée pour tenir compte de ce que la viande stérilisée en boîte est un produit tout à fait différent de la viande congelée.

Dans le cas des petits pois, on constate que si la durée de stockage n'est pas excessive, la congélation est une méthode de conservation moins coûteuse que la stérilisation par la chaleur. Si la durée de stockage est longue, il est au contraire plus économique de conserver les petits pois en boîtes stérilisés que de les congeler. Ce calcul montre qu'il existe une durée de stockage de part et d'autre de laquelle l'un des deux procédés est moins onéreux que l'autre. Cette durée de stockage varie avec chaque denrée et doit faire l'objet d'un calcul distinct pour chaque produit.

The authors give some information concerning cold storage charges in France and some other countries in Europe.

In order to give some idea of the relative costs of preserving foods by heat processing and by freezing, the authors have chosen the examples of meat and green peas. In the analysis made, account has been taken of the costs of processing, storage, transport and distribution of the foods concerned.

In the case of meat, it is difficult to make a valid comparison since the finished products (canned meat and frozen meat) are so different.

In the case of peas where a valid comparison may be made, it appears that, when the time of storage is short, freezing is a cheaper method of preservation. However, as the length of storage increases, this advantage is lost and, when the storage time is long, freezing becomes the more expensive form of preservation. The authors emphasize that the economics of the two processes cannot be judged by a general analysis—a separate calculation must be made for each individual case.

L'HOMME dispose aujourd'hui d'un certain nombre de procédés lui permettant de conserver pendant plusieurs mois une production agricole saisonnière.

Ces procédés font appel à des techniques variées: congélation, stérilisation, dessiccation, concentration en sucre, fumage, etc.—

Au moment où les progrès réalisés dans le

* Ingénieur en Chef du Génie Rural, Chef de la Section Technique du Froid.
† Ingénieur du Génie Rural.



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domaine des radiations annoncent l'apparition d'un nouveau procédé il a paru intéressant de faire le point du coût de ceux qui sont employés aujourd'hui de façon courante. Notre propos se limite cependant aux méthodes qui utilisent le froid et la chaleur.

(1) A savoir pour le froid : la réfrigération et la congélation.
(2) pour la chaleur : la stérilisation ou appertisation.

## COÛT DU SERVICE FROID

La production du froid étant considérée comme une prestation de service, les industriels exploitant des installations frigorifiques ne sont généralement pas propriétaires des marchandises qu'ils traitent. Contrairement aux conserveurs qui interviennent dans le circuit commercial des denrées appertisées, les frigoristes louent seulement leurs services à des tarifs déterminés.

*Tarification française*

La tarification française distingue les frais occasionnés par les manutentions (entrée et sortie des marchandises, pesée, arrimages particuliers etc.) et ceux correspondant au service froid proprement dit à savoir l'entreposage, la réfrigération et la congélation.

Les tarifs de congélation varient avec la température à atteindre. Plusieurs bandes de température sont prévues entre −2 et −30°C. L'opération est facturée à la tonne brute à un prix qui diffère suivant que la congélation a lieu ou non dans la chambre de stockage et qu'elle est opérée sur un produit réfrigéré au préalable ou non.

Les frais de réfrigération et d'entreposage peuvent être calculés à la tonne jour ou à la tonne semaine. Quelquefois, un forfait est perçu pour les 4 premières semaines d'entreposage. Les tarifs dépendent de la température d'entreposage. Cinq bandes de température sont prévues à savoir:

| | |
|---|---|
| Bandes A | Température supérieure à −2°C |
| B | Température comprise entre −2°C et −9°C |
| C | −10°C et −17°C |
| D | −18°C et −24°C |
| E | −25°C et −30°C |

La bande A correspond à l'entreposage des produits réfrigérés, les autres bandes à celui de produits congelés, les bandes C et D étant les plus courantes.

Le tarif de stockage en bande A est de 98 francs par tonne jour. Ce prix comprendrait:

(1) 25 pour cent de charges d'amortissement
(2) 56 pour cent de frais d'exploitation
(3) 19 pour cent de frais généraux.

Le tarif des bandes B, C, D, E est calculé à partir du précédent. Le calcul tient compte du renforcement de l'isolation, de la durée de fonctionnement plus longue des machines et de l'énergie électrique supplémentaire que nécessite la production des frigories à température plus basse.

Le Tableau 1 qui suit indique pour les différentes bandes de température les coefficients de majoration du tarif de base et les tarifs pratiqués.

TABLEAU 1

| | Bande A | Bande B | Bande C | Bande D | Bande E |
|---|---|---|---|---|---|
| Coefficient de majoration | 1 | 1,135 | 1,35 | 1,63 | 2,065 |
| Tarif | 98 | 110 | 140 | 170 | 200 |

*Tarifications française et étrangères*

Nous reproduisons en annexe un tableau (Tableau 2) qui compare les tarifs d'entreposage pratiqués en France et à l'Etranger. Ce tableau résume les conclusions d'une étude entreprise par les Entrepôts Frigorifiques Lyonnais à l'initiative de leur Directeur Général M. VIDAL qui participe aux travaux de notre session.

Les chiffres de ce tableau ont été calculés en 1957. Ils correspondent à des francs français. Les taux de conversion des

TABLEAU 2. Tableau comparatif des tarifs d'entreposage pratiqués à l'étranger et en France en 1957

Réfrigération

| Denrées | Suisse 81 | France 1 | Italie 0,56 | Hollande 94 | U.S.A. 350 | Angleterre 987 |
|---|---|---|---|---|---|---|
| Viande-la T.–1° mois | 3 081 | 4 089 | 3 080 | 4 610 | — | 5 231 |
| Fruits T.–1° mois | 2 736 | 4 089 | 1 960 | 2 590 | 3 871 | 3 751 |
| Fromages T.–1° mois | 2 736 | 4 089 | 1 960 | — | 8 183 | 4 244 |
| Oeufs-la caisse de 720 unités | 109 | 396 | 173 | 196 | 366 | 197 |

Congélation

| Denrées | Suisse | France | Italie | Hollande | U.S.A. | Angleterre |
|---|---|---|---|---|---|---|
| Viande-la T.–1° mois | 4 215 | 8 152 | 6 440 | 4 610 | 9 800 | 6 317 |
| Fruits T.–1° mois | 4 152 | 9 146 | 6 440 | 8 880 | 7 259 | 6 317 |
| Oeufs T.–1° mois | 4 152 | 6 921 | 6 720 | — | 7 721 | 3 652 |
| Poisson T.–1° mois | 4 152 | 6 921 | 7 840 | — | 9 415 | 6 317 |
| Beurre T.–1° mois | 4 152 | 6 921 | 3 640 | 2 820 | 6 181 | 4 292 |
| Volaille T.–1° mois | 4 215 | 6 921 | — | 4 120 | 8 260 | 6 317 |

N.B. Les chiffres portés sous chaque pays correspondent aux cours qui ont servi à l'étude comparative. Tous les montants sont libellés en francs français.

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monnaies en vigueur à l'époque sont rappelés sous le nom de chaque pays.

Précisons que les tarifs français sont inférieurs de 20 pour cent environ à ceux qui sont en vigueur aujourd'hui, une majoration étant intervenue le 1er janvier 1958.

## COMPARAISON DU COÛT DE LA CONSERVATION PAR LE FROID ET PAR LE CHAUD

*Considérations préalables*

Comparer le coût de la conservation d'un aliment par congélation rapide et stérilisation est une entreprise délicate. En dehors des intérêts commerciaux que cette étude met en jeu, il faut reconnaître que le choix du produit à traiter soulève plusieurs difficultés.

Premier écueil. Certaines denrées soumises à la congélation rapide ne sont pas stérilisées et inversement. Par exemple on conserve le poisson réfrigéré ou congelé. On ne le stérilise pas.

On congèle le beurre mais on ne le conserve pas par la chaleur.

Deuxième écueil. Le produit traité par le froid et par la chaleur doit se présenter au terme de sa conservation sous une forme comestible identique. Par exemple, on conserve la viande à l'état congelé ou dans des boîtes stérilisées. Mais dans un cas la viande est crue, semblable à ce qu'elle était avant traitement; le coût des opérations effectuées représente les seuls frais de la conservation. Dans le second cas, au contraire, on a affaire à un produit fini, en quelque sorte à un plat cuisiné. Les frais engagés ne se rapportent donc pas seulement à la conservation de la viande mais aussi à son assaisonnement et à sa préparation.

Enfin supposant choisi l'aliment à conserver, il importe que les prix de revient cités correspondent à des productions de tonnage voisin.

En définitive quelles que soient les critiques que notre choix peut soulever, nous nous proposons d'examiner les prix de

revient de la conservation de petits pois et de viandes en quartiers.

Nous avons choisi les petits pois, parce qu'aujourd'hui ce légume paraît satisfaire le mieux aux différentes exigences que réclame la comparaison économique proposée.

Toutefois, comme les perspectives actuelles ne laissent pas prévoir l'emploi prochain des radiations pour cette denrée et puisque cette application des rayonnements constitue notre principale préoccupation, nous avons jugé utile d'étudier aussi le coût de conservation de la viande, en dépit des objections formulées plus haut à propos de ce produit.

Il eût été intéressant d'examiner le cas des pommes de terre car les résultats des recherches déjà entreprises permettent de penser que, dans peu de temps, on irradiera industriellement ce tubercule pour l'empêcher de germer. Mais si on peut freiner la germination en ayant recours au froid, il n'existe aucun procédé faisant appel à la chaleur. Disons donc seulement en passant que l'emploi du froid dans ce cas particulier revient à peu près de 2 à 3 Fr/kg/mois.

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*Coût de la conservation des petits pois*

Nous distinguerons les frais occasionnés par:

(1) la fabrication (stérilisation ou congélation)
(2) l'entreposage
(3) le transport
(4) la vente de la marchandise.

*Fabrication.* Nous comparons ici les frais entraînés par la fabrication d'un kilogramme de petits pois conservés.

soit en plaques congelées de 2 kg avec groupage en cartons de 10 plaques;

soit en boites 4/4 de 1 l. contenant 570 g de petits pois stérilisés avec emballage des boîtes en caisses de carton.

En vrac le prix d'achat moyen des petits pois pour la campagne 1958 s'est élevé à 51 Fr/kg. Après battage, triage, transport, le kilogramme de petits pois revient à 66 Fr.

Le coût de la stérilisation s'établit comme suit:

| | |
|---|---|
| Matières premières | 66 Fr |
| Fabrication: | |
| Main d'oeuvre | 18,4 |
| Boîte et caisse | 53,08 |
| Charbon, électricité | 2,63 |
| Sel | 0,47 |
| Frais généraux | 15,62 |
| | % 90,20 Fr |
| Prix de revient industriel | 156,20 Fr/kg |

Le coût de la congélation est le suivant:

| | |
|---|---|
| Matière première | 66 Fr |
| Fabrication: | |
| Blanchiment environ | 5 |
| Emballage fourniture | 15 |
| Emballage main d'oeuvre | 35 |
| Congélation rapide | 9,15 |
| | % 64,15 Fr |
| Prix de revient industriel | 130,15 Fr/kg |

A la sortie de l'usine, les petits pois congelés coûteraient donc environ de 15 à 20 pour cent de moins que ceux mis en boîtes.

Les frais de fabrication sont de l'ordre de grandeur du prix d'achat de la marchandise.

*Entreposage.* L'entreposage des conserves de petits pois en boîtes est opéré par les soins des détaillants et non des conserveurs. Cet entreposage ayant lieu en petites quantités, nous considérerons que les frais correspondant sont négligeables vis-à-vis de ceux qu'entraîne le stockage de petits pois congelés.

Pour ceux-ci, le stockage à $-18°C$ coûte 163 Fr/tonne/jour soit environ 5 Fr/kg/mois. Le prix de revient de la marchandise est donc majoré de 25 pour cent environ après 6 mois de stockage. Le prix de la congélation étant inférieur de 26,20 Fr à celui de la stérilisation, il en résulte que pour un stockage de moins de 5 mois les petits pois congelés reviennent à meilleur compte que les petits pois stérilisés. Au delà de 5 mois, la situation s'inverserait. En réalité, compte tenu des frais de transport et de

vente examinés plus bas, ce délai est probablement ramené à 3 mois. Il s'agit évidemment d'un délai tout à fait approximatif et moyen car les frais de transport par exemple varient avec la distance parcourue.

*Transport.* Pour un parcours de 400 à 500 km environ, le transport de 10 tonnes de petits pois congelés en wagons à isolation renforcée et chargés de 200 kg de glace carbonique revient à 8,37 Fr/kg.

Le transport de conserves de petits pois en wagons S.N.C.F., coûterait pour le même parcours 3,50 Fr/kg. Dans un cas, le transport coûte donc 2,40 fois plus que dans le second. Compte tenu de ce que le chargement comprend aussi le poids de l'emballage, on peut estimer que le transport de petits pois congelés est au moins 3 fois plus onéreux que celui de petits pois en conserves, à poids net égal.

*Vente au détail.* Les frais de vente d'un produit congelé sont plus importants que ceux de produits stérilisés. La différence représente l'amortissement de la vitrine à basse température et la dépense de courant électrique nécessaire à son fonctionnement. Nous évaluons cette différence à environ 4 Fr/kg de marchandises.

*Coût de la conservation de viandes*

Nous comparons ici la dépense qu'entraîne la conservation d'un quintal de viande en quartiers lorsque celle-ci est:

—congelée en caisses après désossage.
—stérilisée sous forme de boeuf en sauce en boîtes de 1 l.
—stérilisée sous forme de boeuf assaisonné en boîtes 1/6 l.

Les résultats de cette comparaison doivent être interprétes en tenant compte de ce que les produits obtenus par ces trois méthodes ne se ressemblent pas au point de vue commercial et que la viande congelée ne provient pas des mêmes parties de la bête que celle qui est stérilisée. Les prix de la viande en font foi.

*Boeuf désossé congelé en caisses*

Prix d'un quintal de viande en quartier 38 150 Fr
Rendement après désossage 73,5%

| | |
|---|---|
| Emballage et conditionnement | 955 Fr |
| Congélation | 514 Fr |
| Main d'oeuvre | 882 Fr |
| Divers frais généraux | 1 062 Fr |
| Total | 3 413 Fr |

Soit 46,50 Fr/kg de viande désossée.

Après un stockage de 6 mois à —18,° le prix de revient de la congélation est porté de 46,50 Fr à 73,50 Fr environ.

*Boeuf en sauce: boîtes de 1 litre*

Prix d'un quintal de viande en quartier 29 700 Fr
Rendement après désossage et parage 62%
Nombre de boîtes obtenues: 79 contenant 546 g de viande chacune.

| | |
|---|---|
| Emballage et conditionnement | 3 190 Fr |
| Fabrication | 5 606 Fr |
| Divers | 1 531 Fr |
| Total | 10 327 Fr |

Le traitement revient donc à 240 Fr/kg de viande comestible.

*Boeuf assaisonné: boîtes de 1/6 litre*

Prix d'un quintal de viande en quartier 26 450 Fr
Rendement après désossage et parage 60,5%
Nombre de boîtes obtenues: 448 contenant 88 g de viande chacune:

| | |
|---|---|
| Emballage et conditionnement | 7 168 Fr |
| Fabrication | 6 097 Fr |
| Divers | 672 Fr |
| | 13 937 Fr |

Le traitement revient à 353 Fr/kg de viande comestible.

Ces exemples montrent d'abord l'incidence des dimensions de l'emballage sur le prix de fabrication. Pour la stérilisation par exemple la dépense est majorée de 47% lorsque le volume des boîtes est réduit de 1 litre à 1/6 l.

En ce qui concerne la congélation, il ressort que la dépense à prévoir varie considérablement avec la durée de stockage de viande en caisses, 10 mois de stockage à —18°C doublent le prix de revient de la congélation.

Nous ne reproduisons pas les chiffres cités à propos du transport des petits pois. Ils s'appliquent sans modifications au cas de la viande.

## CONCLUSION

Nous avons examiné le prix de revient de la conservation par le froid et par le chaud de petits pois et de viande de boeuf.

Les chiffres donnés pour la viande ne sont pas comparables car il s'agit en définitive de marchandises différentes. Ils gardent cependant leur valeur dans la mesure où ils indiquent le coût d'une fabrication déterminée.

Les résultats obtenus pour les petits pois sont plus instructifs. Ils permettent de constater que vraisemblablement il existe un délai de stockage de part et d'autre duquel un des deux procédés de conservation est moins onéreux que l'autre; mais ce délai varie dans chaque cas particulier et sa détermination doit faire l'objet d'une étude plus complète que celle que nous avons présentée.

Dans cet exposé nous nous sommes volontairement bornés à des considérations financières. Sans doute, le point de vue économique est-il essentiel pour permettre à un procédé de conservation d'entrer dans la pratique industrielle, car sa mise en oeuvre doit procurer le gagne pain de ceux qui s'y consacrent. Mais en dessous d'un certain seuil de rentabilité, cet aspect du problème ne doit pas être seul envisagé. Il en est d'autres qui sont au moins aussi importants et qui tiennent à la valeur alimentaire des produits obtenus. Il importe de ne pas les négliger.

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# Costs of Chemical Methods of Controlling Insects in Stored Produce

E. E. TURTLE
Ministry of Agriculture, Fisheries and Food, Infestation Control Division,
Hook Rise, Tolworth, Surrey

Contact insecticides and fumigants are the two main classes of chemicals used for controlling insects in stored products. After a brief survey of present day usage of these techniques, some figures are given for the costs of typical control operations under practical conditions. The object of the paper is to provide a basis on which to compare the potential costs of using ionizing radiations in this field.

In attempting to arrive at a basis for comparing the economics of chemical methods with any new technique, it is desirable to survey, firstly, the types of chemical methods available and, secondly, the conditions under which they are or could be used. Broadly, the chemical methods fall into two main classes of:

(1) contact insecticides, and
(2) gaseous insecticides or fumigants.

Just as the overall usage of the methods varies, the relative extents of the usage of the two classes vary from one country to another.

## CONTACT INSECTICIDES

Insecticides in this class are normally applied as dusts or liquid sprays. The active ingredients include a range of natural and synthetic materials; examples being, pyrethrum extract and DDT, methoxychlor, $\gamma$-BHC and malathion. Most of the contact insecticide used against insects of stored produce is applied to walls of stores, to empty wagons and to outsides of stacks of goods and in similar ways in which the object is to protect clean goods and to keep them free from infestation. There are various reasons, quite apart from costs, why the admixture of contact insecticides is not widely practised. For example, technical difficulties may be encountered in achieving admixture of dust or sprays at an even rate with the commodity. (This is more difficult with some commodities, e.g. dried fruits, than with others, e.g. grain.) Furthermore, many of the contact insecticides are objectionable, either because of residues (e.g. DDT) because of taints, or because of other changes in the qualities of the food. In certain parts of the world inert dusts (e.g. siliceous dusts) are added to grain to protect it from insects. This is done mainly by peasant peoples. Dusts based on pyrethrum or on malathion are also used by admixture to some extent in Europe and America.

## FUMIGANTS

The principal advantage of fumigants is that they enable insects inside packets, inside individual grains and even inside large tonnages of grain and similar commodities, to be killed. The other advantage is that the residues from their use tend to be lower

and less objectionable than when contact insecticides are used. Fumigants are, in fact, used in one form or another in very many countries and with a wide range of commodities.

An analysis of the overall costs involved in fumigating a particular parcel of goods is liable to show that the main costs are those involved in moving the goods to the fumigation chambers, lighters or other site where the fumigation can be carried out. This is particularly marked when the tonnage is heavy. For bagged cereals in country stores for example we have found that the costs of transferring the goods could be ten times as much as the costs of the fumigation itself. To keep costs to a minimum, therefore, it is desirable either to introduce the process as a regular part of the normal chain of storage and distribution, or to develop techniques which can be employed as occasion demands in the various locations where the commodities are normally stored or handled.

Practical developments in fumigation have tended towards the latter course and, at least in the developed and primarily consumer countries, most of the fumigating of basic commodities (grain, oilseeds, etc.) is undertaken by specialized operators. Each fumigation unit usually undertakes treatments in a variety of situations (silo bins, lighters, country stores, bulk floor storage warehouses, etc.) The equipment for each such unit (tools, piping, sheets, vaporizers, etc.) is not highly expensive and many of the items can be used in the various situations. This form of organization, no doubt, is partly because the more highly developed countries receive commodities from many places and because the commodities themselves are not infested sufficiently constantly or not always sufficiently heavily to support the cost of regular disinfestation i.e. as a matter of routine on all of the particular material in commerce.

Even if goods were to be disinfested as a matter of routine (e.g. by irradiation) before going into store it should be borne in mind that they may well become infested during storage. If this situation arises almost any method of treatment *in situ* is likely to be cheaper than the temporary removal of the goods for treatment.

TABLE 1. Tonnages of stored foodstuffs fumigated in various sites

| Site | Tonnage (tons) |
|---|---|
| Under gas-proof sheets | 485,000 |
| In lighters | 215,000 |
| In silo bins | 80,000 |
| In fumigation chambers | 40,000 |
| In buildings | 25,000 |

Table 1 summarizes the situations in which fumigations were carried out on government owned stocks of various foodstuffs in the United Kingdom in 1953.

These figures illustrate the latter point to the extent that most of the fumigations under gas-proof sheets were on stocks which became infested in country stores into which they entered apparently in an uninfested condition.

Although certain commodities are fumigated as a matter of routine in various countries (e.g. dried fruits arriving in the United Kingdom from Australia), the tonnages dealt with are small compared with the tonnages of grain, oil seeds and animal feedingstuffs passing into international commerce. Where grain is concerned the nearest approach to a comprehensive disinfestation service is the existence of apparatus for gassing bins in many silos, e.g. in many ports in Western Europe. The additional cost of installing this gear, when building a new silo, is relatively small and, therefore, no substantial losses are incurred if, as happens in practice, the plant is only used on an intermittent basis.

In my opinion, the best locations for installations for regular treatments are in the producing countries. The producer countries can usually canalize and export their commodities through a limited number of ports and handling installations and if they are situated in tropical or sub-tropical areas their produce is likely to be constantly infested. Examples are: oil seeds and cereals from parts of Africa and South America.

## COSTS

Although it is difficult to give figures for the overall costs of treatments so as to include such incidental charges as those for delaying the movement of goods or taking them to a special station for treatment it is possible to give some figures for the chemical treatments themselves. For this purpose I have chosen examples from Departmental records of the costs of treatment of Government-owned stocks of certain foodstuffs in the United Kingdom. Most of the figures chosen are for the year 1953, because this happens to be a year for which we have fairly comprehensive records.

The figures are approximate and, naturally, they apply only to our conditions. However, they should provide a comparison for the development of ideas about alternative techniques.

*Example* 1. *Fumigation of commodities with methyl bromide in lighters*

These figures were obtained from 88 treatments involving 108,600 tons of cereals, nuts, cocoa and rice, in London in 1953.

| | *Cost per ton* |
|---|---|
| Methyl bromide | 6d. to 1s. 0d. |
| Application and supervision | 9d. to 1s. 6d. |
| Hire of lighters | 4s. to 8s. 0d. |
| Total cost | 5s. 3d. to 10s. 6d. |

Information about similar fumigations carried out in Tanganyika and in Yugoslavia give costs at up to about 50 per cent in excess of these figures.

*Example* 2. *Fumigation of commodities in bags under gas proof sheets in the United Kingdom in* 1953

These figures are based on information about 21 fumigations involving 25,500 tons of cereals.

| | *Cost per ton* |
|---|---|
| Methyl bromide and its application | 2s. 0d. |
| Covering stacks with sheets | 1s. 3d. |
| Deterioration of sheets | 1s. 0d. |
| Total cost | 4s. 3d. |

Figures from the U.S.A. and from East Africa reach up to about 8s. per ton. For fumigating bagged grain with carbon disulphide in Morocco, a figure of 6s. 8d. per ton was quoted commercially.

*Example* 3. *Grain in silo bins*

Various techniques are available and their costs depend largely on whether the silos are equipped with apparatus for gas circulation and whether it is necessary to turn the grain to enable the treatment to take place. This is illustrated by Table 2.

Although these figures are far from precise or comprehensive, I trust that they will be of value when considering the economic potentialities of the use of radiation in this field.

TABLE 2. Costs (per ton) of fumigating grain in silos

| | Calcium cyanide | $CCl_4$ mixture | Aluminium phosphide | MeBr in circulation plant | Pyrethrum PB dust |
|---|---|---|---|---|---|
| Insecticide and application | 1s. 7d. | 9d. | 1s. 6d. | 8d. | 3s. |
| Turning | 8d. | None | 1s. 0d. | None or 1s. | None or 1s. |
| Total cost | 2s. 3d. | 9d. | 2s. 6d. | 8d. to 1s. 8d. depending on type of installation | 4s. 6d. |
| Information on which figures based | 16,800 tons in 1951 | 14,700 tons in 1951 | Based on trials in U.K. in 1957 | (Estimate) | Approximate figures from U.S.A. |

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# The Cost of Chemical Methods of Preventing the Sprouting of Potatoes

W. G. BURTON
Ditton Laboratory, Department of Scientific and Industrial Research, Larkfield, Kent

(1) Spraying growing crop with maleic hydrazide 3–5 weeks before death of haulm.

6 lb MH 40 (sodium salt) per acre at 16s. per lb = 96s. per acre.
Cost of spraying with a high volume sprayer delivering 150 gal per acre (depreciation, tractor, man, etc.) = 30s. per acre.
126s. per acre.

Assume yield to be 8–12 tons per acre; then cost is about:

£0·5–0·75 per ton
or $1·4–2·1/1000 kg

(2) Dusting tubers with tetrachlornitrobenzene in an inert filler. Apply at rate of about 10 lb/ton. Means of application involves negligible capital expense.

Cost: £0·6–0·75 per ton
or $1·7–2·1/1000 kg

(2a) If the dust is blown in by main ventilating fan through the ventilating ducts the consumption may be halved. Use of fan is very brief and therefore need not be costed. This method has been tried in Holland.

Cost: £0·3–0·4 per ton
or $0·8–1·1/1000 kg

(3) Dusting tubers with *iso*-propylphenylcarbamate in an inert filler. As (2) but at rate of 2½ lb/ton. Cost of material in bulk is £250/ton ≡ 5s. 6d. per ton of potatoes. When material is broken down into smaller quantities I have taken it to cost about 60 per cent more and the cost of treatment therefore about 9s. per ton, i.e.

£0·45 per ton
or $1·25/1000 kg

(3a) as under (2a)

Cost: £0·25/ton
or $0·7/1000 kg

(4) Ventilating with nonanol

Cost of ventilating (depreciation, interest, electricity, etc.) 9d.–1s. 9d. per ton
Cost of material 2s. 6d.–5s. per ton
Total cost 3s. 3d.–6s. 9d. per ton
i.e. £0·15–0·35 per ton
or $0·42–1·0/1000 kg

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# The Economics of Treating Fish with Antibiotics

J. M. SHEWAN

Torry Research Station, Department of Scientific and Industrial Research,
Torry, Aberdeen, Scotland

FISH can be treated in two ways:

(1) by incorporation of the antibiotic into ice used for chilling the fish, or
(2) by dipping the fish or, more usually the fillets, into a solution of the antibiotic on shore before dispatch to the inland markets.

At present the only major fish-producing country allowing the use of antibiotics is Canada where both chlortetracycline (CTC) and oxytetracycline (OTC) can be employed. So far they are used almost exclusively with fillets in the dip form.

The treatment recommended by the manufacturers is a quick, i.e. few seconds, dip into 10 p.p.m. CTC or OTC at the rate of 15 lb fish per gal of dip. The fish lie on a moving belt which conveys the fillets into and out of the antibiotic solution. Experience has shown, however, that the concentration has usually to be doubled to ensure adequate treatment.

Assuming each gram of antibiotic activity costs 3s. then the cost per lb at 10 p.p.m. would be about ½d. and at 20 p.p.m. 1d. By such a treatment it is believed that the storage life of the fillets as compared with the controls can be extended by from 40 to 100 per cent in the temperature range of 0–10°C.

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# The Economics of Treating Poultry with Antibiotics

D. H. SHRIMPTON

Low Temperature Station for Research in Biochemistry and Biophysics,
Department of Scientific and Industrial Research, Cambridge

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THE poultry industry of the United Kingdom, in so far as it has been concerned with producing meat, is essentially a post-war development and the greatest expansion has taken place in the past two years. The pattern of development has been towards the establishment of integrated groups of breeders hatcherymen, broiler producers, packers and retailers; about 100,000 broilers a week being killed, dressed and packed by the larger groups. American packing station practice has only been partly adopted in these large packing stations in the United Kingdom. For the most part, freshly killed and eviscerated broilers are chilled in slush-ice in a manner similar to that used in the U.S.A.; but after chilling, the carcasses are wrapped and frozen to aid distribution using transport equipped with some refrigerated containers and an increasing number of insulated ones. In general, the frozen eviscerated carcasses are thawed by the retailer before sale to the consumer.

The costs of this procedure per broiler of 2–3 lb eviscerated weight are of the order of 1d. for wrapping and a further 1d. for freezing. However, a variable amount of water is taken up permanently by carcasses during chilling in slush-ice and for a broiler at the present time this is equivalent to a gain of about 3d.

In addition to preservation by freezing it has been suggested that tetracycline antibiotics could be used to extend the shelf life of chilled poultry or of frozen poultry which had been thawed and subsequently stored under chilled conditions. In the U.S.A., where this process is permitted, the antibiotic is added to the tanks of slush-ice which are used for chilling the freshly killed and eviscerated birds, and gives an extension of storage life of 50–100 per cent at chill temperatures. The process is therefore particularly attractive from an economic standpoint, since it not only requires a minimum of change in current practice, but utilizes the one feature of the packing procedure which results in a gain in weight.

Accurate costing of the use of tetracycline antibiotics as poultry preservatives in the United Kingdom is not possible since their use for this purpose is at present prohibited. However, it is unlikely to exceed 1d. per broiler.

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# Economic Considerations

B. S. SCHWEIGERT
American Meat Institute Foundation and Department of Biochemistry,
The University of Chicago, Chicago, Illinois, U.S.A.

It is my opinion that if approval were granted by governmental agencies today in the United States, there would be few, if any, commercial applications of irradiation made immediately by the food industry. It is likely that several selected applications would be considered with $\beta$- or $\gamma$-ray machines when the costs can be more accurately evaluated. These are as follows:

(1) Extending shelf life of certain fresh meat and fruit products kept under refrigeration
(2) the inhibition of sprouting of potatoes
(3) control of insects in grain.

Application of sterilizing dosages even for the most promising products in the civilian market do not appear likely at the present time. When scientific and technological information has been developed sufficiently, we could expect the use of sterilizing dosages to be applied first to selected meat, poultry and sea-food products.

Current cost estimates for irradiation treatment vary from less than 1 cent to 7 cents per lb. Preliminary calculations suggest that $\gamma$-sources such as are located at Idaho Falls may permit irradiation costs of 2 cents per Mrad/lb. Design and development programs for reactors, fission products and $Co^{60}$ are too indefinite at present to evaluate costs or availability.

The experience to be gained with the design, construction and operation of the pilot plant irradiation facilities in California will be of great value in guiding the future applications by the food industries.

Progress in obtaining information on irradiation of foods may seem slow to many administrators. I believe rapid progress has been made in view of the enormous complexities of the problems and the relative lack of scientific information about foods as we now process them. We should proceed in this field with close co-operation among scientists and with cautious optimism.

In spite of the economic limitations referred to, reflection 10 years from now may well reveal that the basic information obtained on foods in connection with studies on irradiation has justified many-fold the modest research funds spent, even though not a single pound of irradiated food was marketed.

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# Discussion

Dr. Powell (U.K.): Is it possible to obtain a uniform dose with an electron accelerator over nearly spherical objects such as potatoes? This is of importance, since the failure to treat all sprouts on the tuber with a preventive dose might stimulate the sprouting of any eyes receiving a lower dose.

Mr. M. C. Crowley-Milling (U.K.): It is assumed that the electron energy is high enough to fulfil the conditions I mentioned for optimum utilization. At such energies, where the thickness of the layer of potatoes is about two-thirds of the maximum electron range, the variation of dose throughout the potatoes should not exceed $\pm 10\%$.

Dr. Powell (U.K.): A throughput of 90 tons of potatoes/hr is completely unrealistic commercially. A more realistic figure is 5 tons/hr, which would increase costs considerably.

Mr. M. C. Crowley-Milling (U.K.): The figures given were merely to illustrate the quantity of material which could be treated by a 5 kW electron machine. If the commercial demands in this instance are for a lower throughput, then a lower power machine would be used. At a Symposium on the Processing of Foods with Ionizing Radiations at Cambridge in September 1958,[1] I showed a design for a 1 kW portable irradiation unit, which could be used to treat potatoes in the field. A maximum of 18 tons/hr could be treated by such a machine, and the estimated cost of treatment was 6s. per ton.

Mr. Oxley (U.K.): It is difficult to believe that 40 per cent efficiency could be attained with a package irradiation system, since Dr. Cornwell was only able to claim 40 per cent efficiency for a grain-treating plant in which three concentric cylinders of grain were used. This must be close to an ideal geometry and would be difficult to simulate with packages. An efficiency of 30 per cent is probably the best attainable with a package system.

Mr. Murray (U.K.): A package irradiator under consideration uses a plane source sandwiched between two layers of packages. Measurements at W. R. L. with various source and package arrays, have indicated that efficiencies of ca. 40 per cent may be attained.

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# 4. NUTRITIVE VALUE AND WHOLESOMENESS OF IRRADIATED FOODS

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# Safety in the Operation of Radiation Sources and Use of Irradiated Foods

H. F. KRAYBILL*
Chemistry Division,
U.S. Army Medical Research and Nutrition Laboratory,
Denver 8, Colorado, U.S.A.

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The utilization of ionizing radiation for the preservation of foods represents an important advance in the field of food processing. Careful evaluation of the wholesomeness of irradiated foods has been made, and work is continuing; but, thus far, these foods have not been found to be toxic to experimental animals or man. In animal feeding experiments, with a broad spectrum of foods tested, longevity, reproduction and lactation performance in general are the same as for animals maintained on diets of unirradiated food. Investigations conducted to the present time with mice and rats have not revealed any evidence as to radiation induced carcinogens.

While some radiation damage to the macronutrients and micronutrients has been noted, the nutritive value for these foods is equivalent to, or in some cases better than, that for heat processed foods. While radiation processing does not effect a net change in metabolizable energy or the biological value of the protein, when compared to raw food, cooking of lima beans shows a slight improvement in the biological value and digestibility of the protein. Some of the fat soluble vitamins have been found to be quite labile to radiation and, consequently, this destructive action on certain vitamins produces certain biological effects in experimental animals when irradiated foods are fed. The destructive effect of radiation on α-tocopherol (vitamin E) has a pronounced effect on reproductive performance in female rats. There is no corollary for this observation to human nutrition, since the requirement for vitamin E in man has not been established. Recent observations have indicated that vitamin K destruction occurs with concomitant hemorrhage in male rats fed irradiated meats in a specific diet. In further elucidation of this phenomenon, current work has also demonstrated that peroxidized oils and cooked meats when fed to rats on a special synthetic diet will also produce hemorrhagic diathesis. A wide variety of irradiated foods, raw or cooked, when fed to rats will not evoke this hemorrhagic syndrome. Because of the variety of foods consumed in the dietary with adequate intake of vitamin K, it is concluded that no hemorrhagic condition should develop in man receiving irradiated diets assuming, of course, that the observations mentioned above even have any parallelism in human nutrition.

Ionizing radiation has been shown to be quite effective in alteration of proteins, with the result that certain foods exhibit reduced allergenic properties. Of clinical significance, also, is the fact that low radiation doses of 14,000 rads effectively interrupt the reproductive cycle of the trichinae present in pork, and thereby eliminate the public health problem of trichinosis.

Under present methods of processing, none of the irradiated foods has induced radioactivity. High energy radiation in the area of 24 MeV has been shown to produce transient and short lived radioactive isotopes. In the event that accelerated electrons with high energies are utilized in radiation treatment of food, the potential hazard of these radioactive isotopes, which are induced, will then be explored.

An excellent safety program in the operation of radiation sources has been achieved. Radioactive contamination of food cans, by γ-irradiation with mixed fission products in the canal water, has been eliminated. Health physics procedures guarantee the safety of the product as to both contamination and induced radioactivity. Careful monitoring of personnel and equipment at the radiation source has resulted in an excellent record for safety.

* FAO Consultant on Microbiology. Present address: Curtiss Wright Corporation, Princeton, New Jersey, U.S.A.

## INTRODUCTION

With the possibility that the processing of foods by ionizing radiation may become more extensive than that by thermal processing, it becomes quite evident that a rigorous program must be established relative to safety in operation of radiation sources and in consumption of irradiated foods. The public health aspects involved in the radiation processing of foods, by either electron beams or $\gamma$-rays, may be considered through evaluation of the following factors in the testing of the process and product for safety.

(1) Safety in the operation of the radiation source.
   (a) Proper irradiation dose and control of radiation dose by adequate dosimetry.
   (b) Adequate precautions as to shielding.
   (c) Prevention and elimination of contamination of product by radioactive isotopes during processing.

(2) Safety in the use or consumption of irradiated foods.
   (a) Establishment of the lack of induced radioactivity at the current radiation doses used in processing.
   (b) Feasibility of working at higher energy levels and the potential hazard, if any, involved through the presence in the food of short half-life isotopes.
   (c) *Toxicity* or *non-toxicity* of the irradiation end products of the food components, viz. fats, proteins and carbohydrates or even the vitamins.
      (i) Toxicity or wholesomeness of the food will be established using several test animals through typical short term subacute toxicity experiments (8 weeks) or long term chronic toxicity experiments (104 weeks).
      (ii) Through long term animal feeding experiments the effect of irradiated foods on growth, reproduction, lactation, life span and cellular enzymes can be properly evaluated.
   (d) *Nutritional adequacy* of radiation processed foods must be assessed and comparisons made with raw foods and foods processed by conventional heat processing methods. Ionizing radiation can produce changes in the molecular structure of the macronutrients (fats, carbohydrates and proteins) and the micronutrients (especially those labile to radiation). Therefore, feeding experiments and biological procedures must be so designed to ascertain any deviations from the normal with respect to nutrient content.
   (e) Alteration in *antigenicity* of proteins should also be explored with the probability that extensive molecular changes or rearrangements occur in food proteins undergoing radiation treatment. Using "primed" animals (guinea pigs), the extent of reduced allergenicity in irradiated proteins can be accurately measured.
   (f) Since irradiated end products must be proven as toxic or non-toxic in animal and human feeding experiments, in similar manner animal feeding experiments must be set up to establish whether or not irradiated foods possess any induced *carcinogens*. Exhaustive investigations must, therefore, be undertaken to ascertain the carcinogenic properties, if any, of irradiated foods. Extracts from foods which contain compounds of known carcinogenic potential can be tested on mice and rats. The techniques used may include feeding, subcutaneous administration and skin painting.
   (g) The effectiveness of ionizing radiation in processing foods from the standpoint of bacteriological safety must be firmly established. Not only must careful consideration be given to the absence of spoilage micro-organisms, but conclusive evidence must be obtained as to absence of pathogenic micro-organisms.
   (h) Parasitological infestation of foods must also be considered, and the adaptation of radiation treatment of foods must be fully explored in the elimination of such public health hazards.

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(i) Since radiation sterilization and/or preservation of foods is an entirely new concept in food preservation, a testing program on wholesomeness must be comprehensive. Since man's experience for many years has been with heat treated, frozen and dehydrated foods, it is inevitable that comparisons in biological properties of irradiated foods with these types of foods will be made.

## RADIOLOGICAL SAFETY IN γ- AND CATHODE RAY STERILIZATION

*Accelerated electron (cathode ray) sources*

A well planned radiological safety program is required in order to provide adequate protection for personnel operating radiation sources utilized in the sterilization of foods. The essential features of such a program are described fully by LEVIN[1] in consideration of the required radiological safety for a sterilization unit based on the use of accelerated electrons.

One must consider that cathode rays, when passing through materials, yield energy through excitation, ionization and radiation processes. Hence, in the radiation process, this electron energy loss is converted into production of X-rays or bremsstrahlung. Then, too, in the operation of an accelerator, the fast electrons may escape through apertures in the shielding, which is essentially the result of extensive scattering of electrons passing through solid material.

For protection of the operators of the accelerator, a radiation survey should be made with the machine operating at full power to determine the amount of radiation in areas adjacent to the machines. A continuous monitoring of the radiation should be effected for measurement of the sterilization dose, and this can best be accomplished by means of an ionization chamber located inside the cathode ray beam target area. For an automatic control of this radiation monitor, a metering device hooked up to a recorder or alarm system could be used, so that any increase in radiation above the desired dose could sound the alarm and simultaneously de-energize the accelerator.

Warning lights and a system of interlocks to the radiation chamber or cell are precautionary measures in the operation of such a facility. A system of continuous monitoring of the personnel must be carried out by requiring all personnel to wear film badges. These badges are checked routinely for determination of amount of radiation exposure. An educational program on radiological safety for all workers is very important.

A medical protection program for all operating personnel is perhaps the most important safeguard, both from the medico–legal standpoint as well as concern for the health of the employee. Comprehensive physical examinations should be given prior to employment at the radiation facility. Base line data can be secured from these initial "physicals" and particularly pertinent are hematology tests. Follow-up examinations should reflect the health status of a worker who may be exposed to low level radiation.

*Gamma radiation facilities*

Of the large volume of foods that are radiation processed in the United States, the bulk of the tons of food so processed are sterilized by γ-radiation sources. One of the largest processors with respect to volume is the Materials Testing Reactor Gamma Irradiation Facility operated by the Phillips Petroleum Company at Idaho Falls, Idaho, U.S.A.[2]

In this Gamma Facility, where water serves as the shielding material, spent fuel elements are used, which are furnished from the Materials Testing Reactor and the Engineering Test Reactor. The swimming pool or canal is 40 × 6 × 17 ft, filled with demineralized water to a depth of 16 ft. The 16-ft water depth provides shielding to the point that at least all radiation emitted from spent fuel sections is stopped or anyway not measurable at the surface of the water.

The spent fuel elements (3 × 3 × 30 in.)

are transported to the canal in the Gamma Facility building in a 23,000 lb lead cask. Under water the fuel elements are removed from the cask, and placed in specific neutron absorber cadmium lined containers which rest on a stainless steel grid work at the bottom of the canal. Long clamp tools or tongs are used to handle the fuel elements, since they are capable of producing intense $\gamma$-fields ($1 \times 10^7$ r/hr each 24 hr after removal from the reactor and after cooling down in 480 hr $1 \times 10^6$ r/hr).

Some contamination of fuel elements remains after transport from the reactor due to the contact with contaminated reactor water. Hence, this contamination lightly contaminates the clean water of the canal. To eliminate this contamination, metal tanks (water columns) are constructed from the bottom of tank to at least 1 ft above the surface of water of the canal. Clean demineralized water is introduced at the top of each tank which fills and overflows continuously. In these tanks at the bottom of the canal, cans of food are $\gamma$-irradiated by the fuel element sections next to the tank and, hence, these cans never come in contact with the canal water.

*Survey for radioactivity contamination on external surface of cans.* Without the protection of the enclosed water columns, the cans of food would be externally contaminated with radioactivity from the canal water and fissionable material from the fuel element sections. However, after radiation processing, each food can is surveyed with a $\beta$–$\gamma$ survey meter and checked by taking "smears" on each can prior to leaving the building. The smear check is accomplished by rubbing a small piece of paper on the surfaces of the food can and then measuring the radioactive count of the paper in a lead shielded counter. In the event contamination is detected on any of the food cans, a soap and water wash with scrubbing solves this problem unless solid objects are encountered. Ultrasonic vibration will eliminate the solid materials impinging on the can surface, but this process has the disadvantage of homogenizing the canned foods.

*Measurement of neutron flux and estimation of the potential of induced radioactivity.* Both the MTR and ETR spent fuel sections are capable of producing $\alpha$-, $\beta$- and $\gamma$-rays. The $\alpha$-particles are stopped by the aluminum cladding of the fuel elements and the $\beta$-rays by the cadmium (neutron absorber) lined aluminum fuel element containers. The $\gamma$-radiation passes through the water, between the fuel elements, the water columns and the canned food in the water columns.

Neutrons produced in the canal are the result of the $\gamma$-rays reacting on the deuterium in the water surrounding the fuel elements. These neutrons, present in the irradiation zone, are very limited in comparison to the intense $\gamma$-fluxes. In order to determine the magnitude of this neutron flux, a gold foil is irradiated in one position of the canal for 57 hr in an average $\gamma$-field of $5{\cdot}2 \times 10^6$ r/hr. This gold foil is now radioactive and, when counted, gives a calculated neutron flux of $8{\cdot}4 \times 10^3$ neutrons/cm$^2$/sec. Neutron radiation must be minimized to eliminate induced activity. Experience at the Gamma Irradiation Facility or the Materials Testing Reactor indicates that a total dose of $1{\cdot}8 \times 10^7$ neutrons/cm$^2$/sec is tolerable.

*Health physics safeguard at Gamma Irradiation Facility.* Continuous monitoring for potential radiation exposure of personnel is conducted. The maximum permissible exposure (MPE) for personnel at the MRT is limited to a harmless level; no more than 60 mrem/day. Operators then could only work in a radiation field that does not exceed 7·5 mr/hr for an 8 hr day. These are, of course, the upper limits of permissible exposure and, if possible, a negligible or negative radiation field is the objective of all operations.

For safeguarding the personnel, the following procedures are followed in this Gamma Facility area:

(1) Each individual must wear a film badge and dosimeter pencil. Daily checks are made on these monitoring devices.

(2) An area radiation monitor is set up in the building which sets off an alarm when the radiation background exceeds 7·5 mrem. In the event the radiation exceeds this level, the radiological safety officer surveys the

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area for origin of radiation and elimination of same.

(3) While dry radioactive materials are not involved in this facility, as a precautionary measure an air monitor type radiation detector is installed in the building which records the amount of radiation measured on the air filter.

(4) Orientation in radiological safety is given in a short course before each employee starts work. Extensive training is given each worker by a qualified Gamma Facility operator before he handles fuel elements.

(5) Prior to removal of equipment or samples from the canal water, a radiological safety officer must measure the radioactivity of these materials before movement elsewhere. Samples in the water columns such as food cans may be removed without this survey; however, they must be surveyed before they leave the building.

(6) In the event canal water becomes accidentally splashed on the floor or pieces of equipment, immediate removal of the contamination must be accomplished by thorough scrubbing with soap and water.

(7) Before leaving the Gamma Facility, each worker must check hands and feet on the electronic hand and foot counter for detection of any radioactive contamination.

## ESTABLISHMENT OF THE LACK OF INDUCED RADIOACTIVITY IN FOODS

One of the major questions raised, not so much by technical people as by the lay public, is the probability of induced radioactivity in radiation processed foods. It is possible to contaminate foods with radioactive isotopes in three ways: by radioactive fallout; by physiological assimilation of radioactivity by animals or plants; by induced radiation. Our attention is confined to the latter.

It has been demonstrated that energies up to 10 MeV will be acceptable, but beyond this limit many of the common elements in foods can become radioactive. A limit of 10 MeV restricts the penetration of an electron beam into a food packet of approximately 1·5 in., or with two sided irradiation about 3·0 in. It would seem desirable, therefore, to develop the use of higher energy levels for greater penetration. Work is in progress at several research installations considering the application of high energy levels up to 24 MeV for processing of foods.

The neutron flux and activation level at several major irradiation facilities has been measured in the United States. In a survey that was made, it was found that 10 to 10,000 neutrons/cm$^2$/sec were obtained. This resulted in net induced activities of a level below that normally present in foods.

Small amounts of induced activity could be easily overlooked since background radiation obscures these measurements. Most irradiated materials show no activity, when tested with ordinary monitors. Using a precise scintillation counter, MEINKE[3] made measurements for induced radioactivity in foods that were exposed to a kilocurie $Co^{60}$ source. It was found, by taking 20-min counts on the irradiated foods, that no detectable radioactivity in 24 elemental food constituents could be measured. While the radiation source used here emitted only 1·33 and 1·13 MeV $\gamma$-rays, the $\gamma$-radiation sources used as gross fission products will have an energy spectrum somewhat higher. The energy spectrum for any of these $\gamma$-sources is considerably below the energy levels necessary for excitation of the elements to induce radioactivity.

It is possible to calculate neutron induced radioactivity in a food from the potassium, phosphorus and sodium levels. Interestingly enough, PETERSON *et al.*[4] found that foods located 1700 ft away from an atomic bomb detonation had an induced radioactivity which was so low as to make the ingestion of these foods no hazard when compared to the presently accepted tolerance doses.

### *Neutron activation of food*

The principal hazard in the induced radioactivity, produced in foods by neutrons, is the irradiation of the various organs of the body after consuming such foods. Consequently, if a particular organ tends to

extract an appreciable concentration of a radioactive isotope, the hazard of this radiation to the organ and a metabolic process will soon become evident. Both size of the organ and its affinity for a particular element determine the hazard associated with a fixed amount of radiation. For example, the thyroid, which weighs about 20 g in man, concentrates a large part of ingested $I^{131}$. The "bone seeking" characteristic of $Sr^{90}$, which approaches radium in its toxic effect, makes this an extremely hazardous radioactive isotope.

The hazard from the radioactivities depends also on the total time required for the body to eliminate this element. Some long lived isotopes are rapidly excreted and, hence, have a short effective half-life.

## RADIATION PROCESSING AND EFFECT ON THE NUTRITIVE VALUE OF THE MACRONUTRIENTS

An appraisal of the nutritive quality of radiation processed foods is best accomplished through measurement of the biological responses of the macronutrients and the micronutrients remaining in the food following such treatment. A more meaningful evaluation is obtained, perhaps, when comparisons are made with the nutritional values for raw, heat processed and radiation processed foods.

*Proteins*

The extensive animal feeding program is designed to test for the presence, in radiation sterilized foods, of toxic compounds that may be formed in the irradiation of fats, carbohydrates and proteins. Although, initially, a minor effort was concentrated on the determination of the nutritive value of the individual macronutrients, more recently there have been major accomplishments relative to the digestibility and biological value of proteins. The biological value of heat and radiation sterilized proteins of beef, milk, peas, lima beans, wheat gluten and corn have been studied by JOHNSON and METTA.[5,6,7] These investigators found no statistically significant difference in digestibility of nitrogen or in biological value of nitrogen between raw and radiation sterilized beef. The beef (raw and irradiated) was incorporated in the diet at a level of 10 per cent protein ($N \times 6{\cdot}25$) on a moisture-free basis. The rest of the diet, consisting of basal diet, was mixed with the wet beef and stored in deep freeze prior to feeding. These diets were fed to rats for a 16-day feeding period (9 days adjustment period and 7 days collection period, the last 14 days on constant food intake). The collection period was followed by 16 days on a standardizing (low nitrogen) diet in order to determine endogenous and metabolic nitrogen excretions. These values are summarized in Table 1.

Similar experiments to those carried out with beef were also conducted on milk proteins by JOHNSON and co-workers.[5,6] The only weakness in these experiments was the substitution of a milk treated at 210°F for 10 min as the raw milk product. This heat-treated milk was used as the reference product for comparison with similar heat processed products (242°F for 14 min) and those radiation sterilized (2·8 Mrad $\gamma$-ray irradiation). These values are shown in Table 2.

There was no statistically significant difference between treatments with regard to digestibility of the nitrogen of milk protein. However, there was a highly significant difference between the biological value of the treated and untreated milk proteins as noted in Table 2. The lowering of biological value in the heat and radiation processed milk protein may be due to loss in cystine or lysine.

The vegetable proteins of lima beans and peas were first investigated by JOHNSON and METTA;[6] more recently, data have been obtained on corn and wheat gluten protein.[7] The values for the digestibility and biological value of pea protein and lima bean protein are shown in Table 3.

The differences in the digestibility and biological value of pea protein as produced

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TABLE 1. Digestion and nitrogen-metabolism data of the raw and radiation-sterilized beef

| | Raw beef | Radiation-sterilized |
|---|---|---|
| Apparent digestibility (%) | 91·8 ± 0·08 | 92·3 ± 0·50 |
| True digestibility (%) | 100·0 | 100·0 |
| Nitrogen balance (mg/g N intake) | 578·5 ± 9·8 | 591·5 ± 21·0 |
| Biological value | 77·99 ± 1·0 | 78·59 ± 1·5 |

Reprinted from a paper by METTA V. C. and JOHNSON B. C.[5]

TABLE 2. Average biological values and coefficients of apparent and true digestibility of protein for the various milk samples

| | Liquid condensed milk | Heat sterilized condensed milk | Radiation sterilized condensed milk |
|---|---|---|---|
| Apparent digestibility | 86·37 | 85·19 | 85·25 |
| True digestibility | 97·85 | 96·67 | 96·85 |
| Biological value | 89·52 | 84·26 | 81·76 |

$$\text{Biological value} = \frac{\text{N utilized}}{\text{N truly absorbed}} \times 100$$

Reprinted from a paper by METTA V. C. and JOHNSON B. C.[5]

TABLE 3. Effect of heat and radiation processing on the nutritive value of lima bean and pea protein

| | Raw | Heat processed | Irradiated |
|---|---|---|---|
| (a) *Pea protein* | | | |
| Apparent digestibility | 83·5 | 83·6 | 82·9 |
| True digestibility | 92·2 | 91·4 | 90·8 |
| Biological value | 58·3 | 57·8 | 50·5 |
| (b) *Lima bean protein* | | | |
| Apparent digestibility | 61·1 | 67·8 | 61·3 |
| True digestibility | 68·1 | 76·6 | 70·0 |
| Biological value | 48·0 | 64·0 | 47·5 |

Reprinted from a paper by METTA V. C. *et al.*[8]

by either heat or irradiation are not significant, other than the decrease in biological value of pea protein as a result of radiation treatment. In the case of lima bean protein, these differences were more noticeable, particularly in the changes in biological value and digestibility produced by heat treatment. Heat processing improved the digestibility of lima bean protein over that of the raw protein by 9 per cent, and also increased the biological value of this protein by 16 per cent (Table 3). The irradiated bean protein was essentially equivalent to that of raw lima bean protein, showing only slight changes in digestibility (2 per cent) and biological value, no change. Hence, no harmful effect on these proteins can be attributed to irradiation.

The significant effect on the biological value of lima bean protein caused by heat

TABLE 4. Effect of heat and radiation processing on the digestibility and biological value of wheat gluten and corn proteins

| | Unprocessed | Heat* processed | Radiation† processed |
|---|---|---|---|
| (a) *Wheat gluten protein* | | | |
| Apparent digestibility | 91·8 | 91·6 | 91·4 |
| True digestibility | 98·7 | 98·5 | 99·1 |
| Biological value | 41·6 | 41·4 | 41·7 |
| (b) *Corn protein* | | | |
| Apparent digestibility | 83·4 | 83·1 | 84·3 |
| True digestibility | 91·1 | 92·4 | 92·9 |
| Biological value | 48·0 | 45·8 | 47·1 |

* Heated 4 min at 15 lb pressure. Reprinted from a paper by METTA V. C. and JOHNSON B. C.[7]
† Irradiated at 2·8 Mrads.

treatment, increasing the value from 48 to 64 per cent, may be ascribed to the destructive effect on the trypsin inhibitor in lima beans.

Neither radiation sterilization nor heat cooking affects the digestibility of corn and wheat gluten protein, as seen in Table 4.

The biological values of corn and wheat gluten protein are not significantly affected by irradiation or by heat. Since lysine is a limiting amino acid, assays were made on the amino acid in each product at two irradiation doses of 2·8 and 10 Mrads. No significant change in this amino acid was noted due to irradiation treatment.

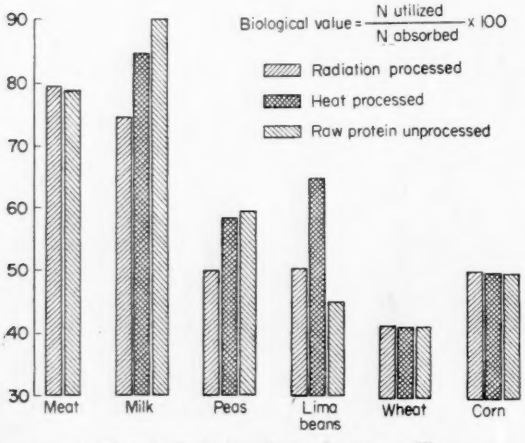


FIG. 1. The biological values of meat, milk, pea, lima bean, wheat and corn protein as affected by heat and radiation processing.

A summarization of the effect of heat and radiation processing on the biological value of proteins is given in Fig. 1.

*Carbohydrates*

Carbohydrates undergo degradative changes similar to proteins when exposed to ionizing radiation. Due to the presence of oxidizing radicals, some splitting of the carbohydrate molecule and subsequent combination of smaller fragments or synthesis (polymerization or cross linking) occurs. The starches and celluloses in foods are of primary interest. Large molecules such as cellulose can be depolymerized by irradiation. These larger carbohydrate molecules can be degraded to glucose which, of course, is readily metabolized *in vitro*. Since amino acids are present in the food with hexoses, such as glucose, it can be expected that some combination (or the browning reaction) will take place between the sugars and the amino acids. Experimental work has shown that this actually occurs just as browning reaction takes place in heat treated foods.

Since carbohydrates supply a major portion of energy in the diet, experiments are usually conducted to determine the irradiation effects on food energy. Since all three macronutrients: fats, proteins and carbohydrates, make a contribution to the metabolizable energy of a diet, the irradiation effect on food energy is not strictly that of its effect on carbohydrate. METTA and

MITCHELL[9] have developed a procedure where the simultaneous determination of the contribution of each dietary component to the metabolizable energy of a ration is made. A multiple regression equation is used to calculate the metabolizable energy. The equation is as follows: $ME = cC + fF + pP$. $C$, $F$ and $P$ are the amounts of carbohydrate, fat and protein consumed, and $c$, $f$ and $p$ are constants determined from collected sets of data by the method of least squares. This method represents the metabolizable energy (the energy digested and absorbed which is utilized by animal and not excreted) per gram of the respective nutrients.

The metabolizable energy was the same for irradiated and non-irradiated foods. There were no differences observed when either the foods or prepared diets were irradiated or the individual nutrients (fats, carbohydrates, proteins) were irradiated. These observations were made at radiation levels of 3 and 6 Mrads, which is within the range normally used for sterilization of food. Some values for metabolizable energy of the micronutrients are presented in Table 5.

Similar experiments conducted in our laboratory, in which rats were fed a composite diet of nine foods (irradiated and unirradiated) for two years, failed to show any differences in metabolizable energy for these diets, indicating again that radiation had no effect on the food energy value of the foods. Other experiments on our metabolic ward, where 10 human volunteers were fed irradiated and unirradiated foods in their diet for at least 15 days, also failed to show any differences in the ME value for such foods as the result of radiation processing. Some diets, which contain as much as 70% water and which were irradiated to 9·3 Mrads when fed to rats, were completely acceptable and promoted normal gains over a short period of time.

TABLE 5. Metabolizable energy of nutrients

| | Non-irradiated (cal/g) | Irradiated (cal/g) |
|---|---|---|
| Casein | 4·56 ± 0·30 | 4·41 ± 0·22 |
| Lard | 8·82 ± 0·31 | 8·87 ± 0·39 |
| Carbohydrate* | 3·87 ± 0·20 | 3·78 ± 0·30 |

* Sucrose, dextrin and starch (2 : 8 : 1)

Reprinted from a paper by JOHNSON B. C. and METTA V. C.[6]

*Fats*

Extensive studies have been conducted on the radiation effects on triglycerides and fatty acids. While polymerization and oxidation occurs when fats are irradiated, some decarboxylation also takes place. The most interesting effect of irradiation is, however, the increased susceptibility of fats to oxidation, the disappearance of the naturally occurring antioxidants (α-tocopherol or vitamin E) and the formation of peroxides with subsequent decrease in unsaturation. The formation of these peroxides and subsequent degradation to carbonyls (aldehydes and ketones) is, in effect, a measure of the extent of radiation damage to fats. Whether any of these oxidation products formed by irradiation have a toxic or deleterious effect, or serve as antagonists to the vitamins *in vitro*, or reduce the total nutritive value of the fats must be determined by exhaustive animal feeding experiments. Theoretically, it may not be easy to make the distinction, after feeding peroxides or carbonyls (present in irradiated fats), as to whether gross symptoms observed in the animal are the result of a toxicity, or the reflection of a nutrient inadequacy. Essential fatty acids may, in truth, be transformed by irradiation and, hence, may not be available to the animal. The carbonyls which are formed are known at least to destroy some fat soluble vitamins, especially vitamin E, and may act as antagonists to others. The biological responses of animals, and ultimately man, to irradiated fats are, therefore, of much interest.

Oxidized and polymerized fats have been studied for many years as to their physiological effect on the animal. CRAMPTON *et al.*[10] in 1951 showed that polymerized fats are toxic when fed to rats. ANDREWS *et al.*[11] have conducted the most extensive

studies on the biochemical effects of feeding irradiated fats and fatty foods to rats.

Autoxidized soybean oil, separated into a high peroxide fraction (Peroxide No = 3185), an intermediate fraction (PN = 927), and a low peroxide fraction (PN = 13), has been fed to rats. Repeated experiments have shown that the low peroxide fraction had no toxic effect on rats. These animals consumed food well and gained weight. Oxidized oils with peroxide numbers of 927 and 1156 were fed to rats, and did not produce as good a growth in 14 days as the oil with a low peroxide number. The hair coat of the animals fed the higher peroxide fraction oil was somewhat oily and unkempt as compared to those animals on the low PN

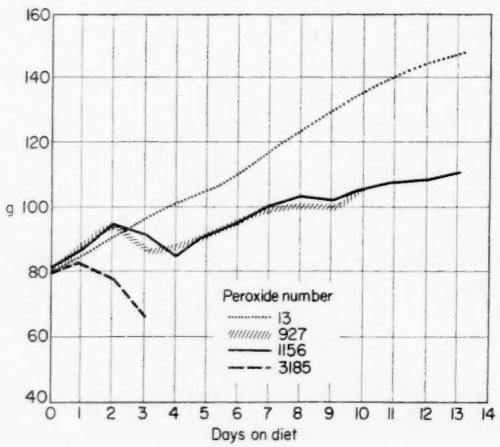


FIG. 2. Growth of rats on various fractions of autoxidized soybean oil. (After J. F. MEAD and W. H. GRIFFITH, by courtesy of the University of California at Los Angeles.)

oil. Another group of animals on an extremely high peroxidized oil (PN = 3185) failed to grow properly, losing weight in the first day and mortalities occurred shortly thereafter (Fig. 2). Histopathological examinations made on the animals failed to reveal any anatomical cause of death. There was some slight enlargement of the liver. From more recent knowledge concerning the effect of carbonyls on vitamin K synthesis *in vivo*, one might suspect that an effect on the blood clotting mechanism should have been evident in this work.

These recent studies by the group at the University of California at Los Angeles are in contrast to their earlier studies where they concluded that 100 PN fat was not toxic, a 400 PN fat caused reduction in growth, and the consumption of a 1200 PN oil diet caused death within 3 weeks. Chief symptoms were loss of weight and diarrhea. Most of the fats in radiation sterilized foods treated at 2·8 or 5·6 Mrads do not have peroxide numbers exceeding 100. It is of interest to note that, in recent work by these same investigators, the level of non-oxidized oils in the diet had no effect on the toxicity of the oxidized lipids. Since some of the toxic effect may be due to disappearance of antioxidants, certain rations were added to a diet to inhibit the oxidation. By adding ferrous and manganese sulfate at concentrations of $10^{-4}$ M, there was a beneficial effect on growth.

ANDREWS *et al.* have also been feeding bacon treated at 2·8 and 5·6 Mrads and, thus far, have not observed any toxic effects from treatment of fats at this radiation dose.

Consistent with previous findings, inhibition of rat intestinal xanthine oxidase by a diet containing 20 per cent of air oxidized soybean oil (PN = 1200) has been observed. Further elucidation of the mechanism of this enzyme inhibition has led to the hypothesis that inhibition of this enzyme is due to some sort of oxidative reaction between flavin adenine denucleotide and oxidized lipids. Earlier experiments also revealed that, in cytochrome oxidase studies, manganous ion curtailed the rate of ascorbate oxidation *in vitro*, but did not exert this effect in the presence of tertiary butyl hydroperoxide.

Repeated experiments conducted in our laboratory have shown an increase in cytochrome oxidase activity in successive generations of rats fed an irradiated composite diet of nine foods, and this effect was attributed to the presence of meat in the diet. This increase was further augmented by room temperature storage of irradiated beef prior to feeding, and an additional increase took place as a result of cooking this product. It is believed that the observed increase in

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cytochrome oxidase activity is related to some oxidative phenomenon, and the increase may be largely manifested through presence of corn oil in diet fed to rats. Further work is required to determine the role of irradiated and non-irradiated fats in relation to this increase in activity of the cellular enzyme.

## RADIATION PROCESSING AND EFFECT ON THE NUTRITIVE VALUE OF THE MICRONUTRIENTS

The radiosensitivity of pure solutions of vitamins varies considerably; the destructive effect of radiation on vitamins in foods differs from that in pure solution. For example, the radiation destruction of vitamin $B_{12}$ in milk at a dose of 46,500 rads is only 30 per cent, whereas in aqueous solutions at the same radiation dose 68 per cent of this vitamin is destroyed. Natural protectors in foods or the presence of other vitamins reduces the amount of radiation destruction. Niacin exerts a protective effect on ascorbic acid, and the presence of oxalic acid increases the radiosensitivity of niacin. Thiamine and ascorbic acid are quite labile to radiation, while niacin and riboflavin are somewhat more radioresistant.[12]

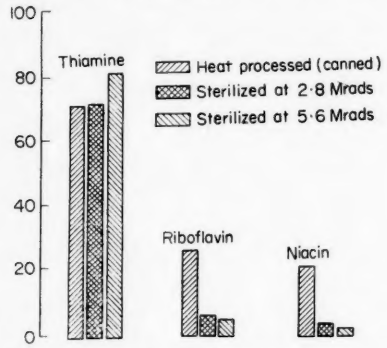


FIG. 3. Destruction in beef processed by heat or $\gamma$-radiation.

In chemical, microbiological and biological (rat growth) procedures, DAY *et al.*[13] found that $\gamma$-radiation of raw ground beef (3 Mrads) resulted in destruction of about 10 per cent of riboflavin and 25 per cent of pyridoxine.

The effect of heat processing and $\gamma$-radiation on the thiamine, riboflavin and niacin content of beef is illustrated in Fig. 3. It is apparent with respect to this one food that radiation processing has no greater destructive effect on these vitamins than the conventional heat processing of foods. In our laboratory five foods were assayed for thiamine, riboflavin and niacin prior to irradiation and again after irradiation at 2·8 and 5·6 Mrads.[14] The greatest destructive effect was on thiamine, Table 6. A rather interesting effect which at first seemed inconsistent was the large percentage destruction of niacin in peaches. Niacin has been considered rather radioresistant, but since ascorbic acid was used in the commercial pack frozen peaches which were irradiated, it must be remembered that

TABLE 6. Vitamin destruction in foods exposed to varying doses of $\gamma$-radiation (% destruction)

| Food | Thiamine | | Riboflavin | | Niacin | |
|---|---|---|---|---|---|---|
| | $2{\cdot}8 \times 10^6$ rads | $5{\cdot}6 \times 10^6$ rads | $2{\cdot}8 \times 10^6$ rads | $5{\cdot}6 \times 10^6$ rads | $2{\cdot}8 \times 10^6$ rads | $5{\cdot}6 \times 10^6$ rads |
| Beef | 76 | 85 | 5 | 4 | 2 | 1 |
| Ham (fresh) | 87 | 96 | 0 | 0 | 2 | 2 |
| Peaches | 94 | 98 | 0 | 0 | 48 | 56 |
| Beets | 52 | 75 | 14 | 10 | 0 | 10 |
| Powdered milk | 0 | 0 | 0 | 0 | 35 | 20 |

Reprinted from a paper by ZIPORIN Z. Z. *et al.*[14]

niacin destruction upon irradiation is intensified in the presence of this vitamin.

In chick feeding experiments, RICHARDSON *et al.*[15] noted that menadione was appreciably destroyed by doses of 2·8 Mrads. The development of hemorrhage was used as the criterion for the vitamin destruction. Vitamins $K_1$ and $K_5$ were not nearly as susceptible to radiation treatment. A synthetic basal ration irradiated at 2·8 Mrads, and fed to chicks, caused an incidence of hemorrhage to the extent of 86 to 95 per cent. By feeding irradiated alfalfa leaf meal or spinach, no destructive effect of vitamin K with resultant hemorrhage took place. Quite recent work, as yet unpublished, has shown that when rats are fed irradiated ground beef in a diet unsupplemented with menadione a significant destruction of vitamin K takes place, as evidenced by the occurrence of hemorrhage after 30 days of feeding. The high percentage of mortalities approaching 80 per cent is restricted more to male weanling rats than to female rats of the same age during the growth period.

The production of this hemorrhagic diathesis in rats fed irradiated beef has been observed with a special diet used by JOHNSON and his colleagues at the University of Illinois. The hemorrhagic syndrome could be corrected by oral administration or intramuscular injection of menadione. Of paramount interest in these investigations is the fact that no coprophagy is practiced by rats maintained on a diet of 5·6 Mrads irradiated beef. This would account for the failure of the rat to secure enough vitamin K from feces by recycling, and from this deficiency a high mortality rate was noted with rats fed this type of beef. The beef treated at a lower radiation dose of 2·8 Mrads does not cause as extensive destruction of vitamin K, and the animals maintained on this diet practice coprophagy to the extent of consuming about 45 per cent of their feces. In consuming a normal basal ration, a rat may recycle as much as 75 per cent of his feces, thereby providing an adequate intake of synthesized vitamin K.

As would be expected, the hemorrhagic diathesis has been produced in rats consuming irradiated pork as 35 per cent of the diet. Carbonyls in irradiated beef have been suspected as the compounds responsible for the inhibition of vitamin K synthesis in the gastrointestinal tract. This assumption has been confirmed by the production of hemorrhage in rats, after feeding highly peroxidized oils and administering microgram quantities of short chain aldehydes. Cooking of irradiated meats further intensifies the formation of carbonyls with the ultimate production of hemorrhage in rats when such foods are fed.

While the production of this hemorrhagic diathesis in rats fed irradiated meats or fats is attributed to an artifact of the diet and the antagonistic action of carbonyls, it is most significant that autoxidation (or heat processing of foods) induces the same effect when employing the same type of experimental diet. A diet consisting of a broader spectrum of irradiated foods without supplementation of menadione presumably will not produce hemorrhage in rats, even if beef is fed in the diet. These studies are being extended to elucidate the mechanism of vitamin K destruction in the gastrointestinal tract of the rat when fed certain irradiated products, or even overcooked or oxidized meats and fats.

Earlier work in this laboratory[16] and elsewhere has shown that vitamin E was readily destroyed in the irradiation of foods. Furthermore, it was also demonstrated that irradiated foods with high peroxide content were capable of destroying vitamin E, either on contact in storage of a diet or by contact with the food in the gut of the rat. This destructive effect on α-tocopherol resulted in reproductive failure with female rats. Vitamin A is also labile to radiation, and as much as 85 per cent destruction of this vitamin has been noted in treatment of milk at 0·44 Mrad.

Although extensive destruction of vitamins may take place by radiation processing of foods, the average extent of this destruction is no greater than that observed in thermal treatment of foods. From practical considerations, then, it is obvious that a varied diet of irradiated foods should provide adequate levels of all vitamins for human consumption.

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## LONG TERM ANIMAL FEEDING EXPERIMENTS (CHRONIC TOXICITY) ON IRRADIATED FOODS AND EFFECT ON REPRODUCTION, LACTATION AND LONGEVITY

Before any release of irradiated foods for general consumption by the public can be made, it must definitely be established that the end products of radiation preservation or sterilization are non-toxic. The procedures for guaranteeing that these foods do not have induced radioactivity have already been discussed. Through an animal feeding or screening program using rats, dogs, mice, chickens and monkeys, it is possible to evaluate the wholesomeness of radiation processed foods. In such an evaluation of wholesomeness, there are two major considerations in long term feeding trials: (a) Potential or chronic toxicity, and (b) nutritional adequacy. The latter subject has already been discussed. The eight or twelve week feeding experiments provide data on subacute toxicity. For chronic toxicity evaluation the animals are usually fed a minimum period of 2 yr, or in the case of rat feeding experiments through four generations of animals.

In addition to securing data on nutritional adequacy and potential toxicity, these feeding experiments provide important information on growth, feed efficiency, reproduction, lactational performance and span of life (or longevity) of an animal maintained on a specific food or diet.

Approximately 44 foods have received toxicity clearance by means of subacute toxicity or short term experiments. An additional 64 irradiated foods have been cleared for human consumption by extrapolation from this basic group of 44 foods. Clearance for human consumption at the moment only provides for taste panel (or palatability) evaluation, and not for continuous consumption of these irradiated foods. Negative toxicity data from the 2-yr feeding experiments, in which rats and dogs are fed radiation processed foods, will ultimately permit the complete clearance for wide scale consumption of these products.

For a significant challenge in testing such foods, the U. S. Food and Drug Administration requires that the test animal receive at least 35 per cent dietary solids of the irradiated food. The remainder of the diet consists of a nutritionally adequate, non-irradiated, semi-synthetic or basal ration. Currently, all irradiated foods have been sterilized at two radiation doses 2·8 and 5·6 Mrads. These irradiated foods treated at both radiation levels are stored a minimum period of 3 months at 70°F prior to feeding. The reference or control diet, which is fed to the test animal, contains an unirradiated raw food. This is stored frozen prior to feeding. Since all animals are randomly selected and distributed into feeding groups: control, X irradiation group (2·8 Mrads), and 2X irradiation group (5·6 Mrads), and

TABLE 7. Listing of twenty-one foods currently under investigation by the Department of the Army, U.S.A.

| Food tested | Test animal | | | Food tested | Test animal | | |
|---|---|---|---|---|---|---|---|
| | Rat | Monkey | Dog | | Rat | Monkey | Dog |
| (1) Beef, Ground | x | | x | (12) Corn | x | | x |
| (2) Pork Loin | x | | x | (13) Beans, Green | x | | x |
| (3) Bacon | x | | x | (14) Potatoes | x | | x |
| (4) Shrimp | x | | x | (15) Potatoes, Sweet | x | | x |
| (5) Codfish | x | | x | (16) Flour | x | | x |
| (6) Chicken | x | | x | (17) Fruit Compote | x | | x |
| (7) Tuna | x | | x | (18) Evaporated Milk | x | | x |
| (8) Beef Stew | x | | x | (19) Peaches | x | x | |
| (9) Chicken Stew | x | | x | (20) Oranges | x | x | |
| (10) Carrots | x | | x | (21) Jam | x | | x |
| (11) Cole Slaw | x | | x | | | | |

TABLE 8. Effect of irradiated diet on the growth and fertility of the albino rat

| | Control diet | Irradiated diet |
|---|---|---|
| Days on diet | 540 | 540 |
| Average final weight, first generation (males) | 430 | 440 |
| Average final weight, first generation (females) | 285 | 295 |
| Total number of litters (four generations) | 105 | 66 |
| Number of sterile males (four generations) | 1 | 6 |
| Total number of living males (all generations) | 293 | 140 |
| Total number of living females (all generations) | 304 | 180 |

Reprinted from a paper by DACOSTA E. and LEVENSON S. M.[16]

fed the same percentage of food in the diet, comparisons between the responses of the irradiated and unirradiated diet can be made.

A basic group of 21 irradiated foods common to civilian and military dietaries has been selected, and is currently under investigation in a 2-yr testing program with rats, dogs and monkeys. A listing of these foods and the test animal used for these toxicity experiments is presented in Table 7.

Perhaps the first long term feeding of irradiated foods was that reported by DA COSTA and LEVENSON.[16] They studied the effect of feeding irradiated semi-synthetic diets to rats. The results of these experiments are shown in Table 8.

The loss in fertility and decrease in life span was attributed to vitamin E destruction. These findings were later confirmed by POLING *et al.*[17] at Swift and Company in a 2-yr feeding experiment on 2 Mrads irradiated beef (Table 9). They concluded that "treatment of beef muscle with a sterilization dose of radiation of high velocity electrons does not significantly impair the nutritional value or wholesomeness."

Several other investigators encountered the problem of vitamin E destruction in irradiated foods when these products were fed to rats. The general practice at the present time is to furnish a supplement of fat soluble vitamins including vitamins A, D, E and K. The supplement is given to the

TABLE 9. Life span data on rats maintained on irradiated and on non-irradiated beef diet

| | Males | | Females | |
|---|---|---|---|---|
| | Non-Irradiated beef | Irradiated beef | Non-Irradiated beef | Irradiated beef |
| Number of rats started | 40 | 40 | 40 | 40 |
| Deaths during experiment | | | | |
| (a) 0–30 weeks | 0 | 1 | 0 | 0 |
| (b) 31–52 weeks | 0 | 3 | 1 | 1 |
| (c) 53–78 weeks | 8 | 7 | 0 | 7 |
| (d) 79–104 weeks | 17 | 12 | 12 | 13 |
| Total | 25 | 23 | 13 | 21 |
| Animals sacrificed | 10 | 10 | 10 | 10 |
| Animals surviving (104 weeks) | 5 | 7 | 11 | 9 |
| Average days survival | 614 ± 18·8 | 581 ± 24·9 | 685 ± 14·7 | 612 ± 21·8 |

Reprinted from a paper by POLING C. E. *et al.*[17]

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animal orally at a time when contact of irradiated food with the vitamins in the gastrointestinal tract is minimized.

Organ meats which are high in sterols, vitamins and other metabolic intermediates have been irradiated (2·8 Mrads) and fed for 2 yr to rats.[18] A complete supplement of non-irradiated vitamins is furnished. Following four generations of feeding, no adverse effects on growth, reproduction or longevity were noted.

At our laboratory a study has been completed (with exception of histopathology), in which a diet compounded of nine individually irradiated natural food items were fed for 2 yr.[19] All of the foods were irradiated to a dose of 5·6 Mrads. A non-irradiated vitamin supplement was added to all diets. In a comparison of two diets, irradiated and non-irradiated, it was noted that the reproductive and lactational performance of female rats on both diets were equal. No adverse effect on longevity of males and females on either diet was observed (Table 10).

Experiments with rats, dogs and monkeys on the feeding of irradiated foods stored at 70°F for minimum period of 3 months are continuing. None of the data reported thus far on these animal feeding experiments would indicate any significant differences between irradiated and non-irradiated foods relative to growth, reproduction, lactation or life span. Within certain generations, and at certain times during the feeding experiment, some differences between the response of rats on the irradiated and non-irradiated diets appeared noticeable. However, averaging out the results for those experiments now concluded on a few foods, it is definitely shown that these foods are non-toxic and will support normal growth in the animal. Typical growth curves for rats fed room temperature stored irradiated and non-irradiated pork are shown in Fig. 4.

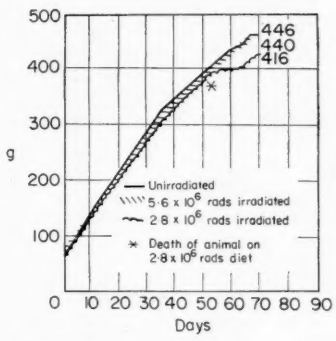


FIG. 4. Growth of male albino rats raised on diets containing irradiated pork stored 3 to 6 months at 70°F.

TABLE 10. Reproduction and lactation data from the first litters of the parent generation raised on composite non-irradiated and irradiated diets

| | Non-Irradiated | | Irradiated | |
|---|---|---|---|---|
| | Number | % | Number | % |
| Number of pups counted | 144 | — | 146 | — |
| Alive | 127 | 88 | 122 | 84 |
| Dead | 17 | 12 | 24 | 16 |
| Total pups born litter | 9·0 | — | 9·7 | — |
| Pups per litter weaned | 6·8 | — | 6·4 | — |
| Average weight (g) for: | | | | |
| Pups born | 6·0 | | 6·1 | |
| Pups weaned | 29·5 | | 29·2 | |

## INVESTIGATION OF POSSIBLE CARCINOGENICITY OF IRRADIATED FOODS

The long term feeding experiment, carried through 2 yr with rats, provides an opportunity to observe any spontaneous tumor formation in animals either consuming

TABLE 11. List of foods, radiation dose, testing technique and test animals used in carcinogenicity studies

| Food | Radiation dose (Mrads) | Testing technique | Test animal |
|---|---|---|---|
| Bacon | 5·58 | Painted, fed | Mouse |
| Beef sterol | 5·58 | Injected, fed, painted | Rat, mouse |
| Yeast sterol | 5·58 | Injected, fed, painted | Rat, mouse |
| Mixture: Corn oil | 0·465 | Injected, painted, fed | Rat, mouse |
| Mixture: Peanut oil | to | Injected, painted, fed | Rat, mouse |
| Mixture: Cottonseed oil | 5·58 | Injected, painted, fed | Rat, mouse |
| Pork | 5·58 | Fed | Mouse |
| Chicken | 5·58 | Fed | Mouse |
| Milk | 5·58 | Fed | Mouse |
| Carrots | 5·58 | Fed | Mouse |
| Potatoes | 0·01 | Fed | Mouse |
| Brain-Egg | 9·30 | Fed, injected | Rat, mouse |
| Mixture: Meat, Fish, Cheese, Milk | 2·79 | Painted, fed | Rat, mouse |
| Mixture: Lard | 5·58 | Injected | Rat, mouse |
| Mixture: Codfish | 5·58 | Fed | Mouse |
| Mixture: Chicken stew | 5·58 | Fed | Mouse |
| Mixture: Beef stew | 5·58 | Fed | Mouse |
| Mixture: Beans, Green | 5·58 | Fed | Mouse |
| Mixture: Peaches | 5·58 | Fed | Mouse |
| Mixture: Flour | 0·074 | Fed | Mouse |
| Mixture: Beef, Tuna, Corn, Sweet potatoes, Fruit compote | 5·58 | Fed | Mouse |

irradiated or raw foods. From theoretical considerations, it was initially assumed that certain compounds in foods, such as the sterols, when irradiated could become potential carcinogens. An extensive testing program is underway at several research institutions to determine the carcinogenic activity of irradiated food products and food components. The test animals used are rats and mice. The foods are either fed to rats and mice, injected, or extracts of the food painted on the skin. A list of foods under test, the radiation dose applied to the food, the experimental technique used and the test animal employed are shown in Table 11.

Thus far, no statistically significant differences in the rate of tumor formation have been observed with either rats or mice which have been tested with irradiated and non-irradiated foods. Most of the investigations, however, have not progressed far enough to establish definite conclusions.

## EFFECT OF FEEDING IRRADIATED FOODS ON TISSUE AND BLOOD ENZYME LEVELS IN THE RAT

Reference was made in previous discussion on feeding irradiated meats and fats, that slight elevations were observed in activity of liver cytochrome oxidase and intestinal xanthine oxidase when rats were fed irradiated foods. Other experiments have shown that, within the third generation of rats, a slight elevation of liver, heart and kidney cytochrome oxidase was noted when irradiated pork was fed. In the same experiment, the preceding generations of rats failed to reveal any significant differences between tissue enzyme levels for animals on either irradiated or non-irradiated diet. Liver choline oxidase activity from animals (third generation) on a control diet was slightly

higher than the value for animals on an irradiated diet.

In the *in vivo* studies, the following enzyme systems have been applied to this work: (a) cytochrome oxidase, (b) choline oxidase, (c) DPNH oxidase, (d) oxidative phosphorylation enzyme, (e) xanthine oxidase, (f) succinoxidase, (g) liver and serum alkaline phosphatase, (h) hypotonic endogenous and (i) serum glutamic-pyruvic transaminase. In the *in vitro* studies on irradiated fats, carbohydrates and proteins, chymotrypsin, trypsin, pepsin, α-amylase, sucrase and lipase were utilized to detect any differences in the digestibility of the irradiated and unirradiated macronutrients by any of the listed enzymes.

It was noted in our laboratory that, when a composite diet composed of cooked irradiated foods was fed to growing rats, the activity of serum glutamic-pyruvic transaminase was decreased as compared to the serum enzyme values in rats that were fed either non-irradiated or raw irradiated foods. It is presumed that cooking at least has an effect on pyridoxine, the limiting vitamin, which is reflected in the alteration of serum transaminase.

Several investigators have shown that the *in vitro* digestion of fats is reduced by irradiation of the fat. Irradiated fats stored frozen or at room temperature are also less susceptible to action by lipase.[20] While sucrose in aqueous solution when irradiated was not less susceptible to attack by sucrase, sucrose irradiated in the dry state showed reduction in digestibility by sucrase. Casein irradiated in the dry state showed no decrease in digestibility by chymotrypsin, but casein irradiated in 5 per cent solution resulted in a depressed ability of the casein to be digested by the protease.[20]

## HUMAN VOLUNTEER FEEDING STUDIES ON IRRADIATED FOODS

Forty-two foods irradiated at 2·79 Mrads and stored frozen, which passed exhaustive animal experiments, have been fed for 15-day periods to 10 human volunteers at our laboratory. Of this group, nine of the irradiated foods were less acceptable than the same foods non-irradiated. Balance studies indicated that there was no difference in the metabolizable energy, nitrogen balance or coefficients of digestibility. Room temperature stored irradiated foods have been and are being, fed to the test subjects. Ground pork loin irradiated at 2·79 Mrads and stored for 12 months at 70°F has been fed for a 15-day period at level of 32 per cent of calories in the diet. The acceptability of this product was not as good as the control or unirradiated pork.

In another experiment, eight foods (cod-fish, bacon, fruit compote, chicken, peaches, green beans, flour and potatoes) comprising 80 per cent of the diet were fed for 15 days to the human volunteers. Potatoes and flour were the only irradiated foods (low radiation dose) that were not different from the identical untreated foods. Clinical and laboratory examinations have failed to reveal any significant alterations in the test subjects consuming either irradiated or non-irradiated foods for the 15-day period.

Future studies with human volunteers will be conducted wherein irradiated foods will be fed for extended periods of 6 to 9 months. A large program of feeding irradiated foods to military personnel on an experimental basis is now under way at Fort Lee, Virginia, U.S.A., for the main purpose of evaluating the acceptability of selected irradiated foods.

## PARASITOLOGICAL ASPECTS OF FOOD IRRADIATION

In 1954 Gomberg *et al.*,[21] at the University of Michigan, established the feasibility of controlling trichinosis by irradiation of pork. About 0·7 to 0·9 Mrad are required to kill trichinae *in situ*, but 19,000 rads of $Co^{60}$ γ-rays will stop the development of larvae to adults, and 11,000 rads will sterilize the females, thus preventing reproduction. Both of these radiation doses will effectively control trichinosis. Operating at these low

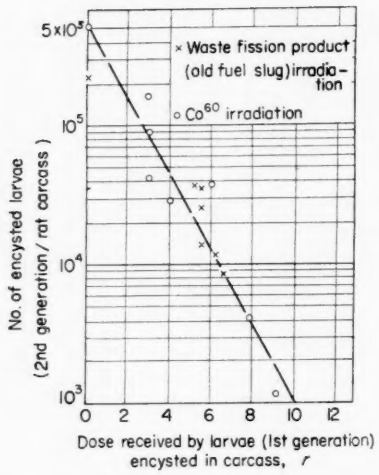
5x10⁵
10⁵
10⁴
10³
× Waste fission product (old fuel slug) irradiation
○ Co⁶⁰ irradiation
No. of encysted larvae (2nd generation / rat carcass)
0 2 4 6 8 10 12
Dose received by larvae (1st generation) encysted in carcass, r

FIG. 5. Larvae in rat muscle 30 days after feeding 5000 irradiated larvae. (Reproduced from a paper by GOMBERG H. J. *et al.*[21])

radiation doses is economical, and the amount of irradiation applied to a hog carcass is not enough to produce the deleterious off-odor and off-flavor changes encountered at the sterilization dose of 2·8 Mrads. Mixed fission products and 200-kV X-rays have also been used effectively in this processing of pork.

Using $\gamma$-rays with energies of 0·6 MeV or greater to irradiate the hog carcass, the dose variation was only 15 per cent. With the irradiation source approximately 13 to 17 in. from the hog carcass, the radiation penetration required was 10 to 14 in. At these distances, the dosimetry varied about 2 to 1 from the surface to the interior of a 14-in. thick section.

Using the $Co^{60}$ source the irradiation was conducted in air, while with mixed fission products the irradiation was conducted in a 20-ft deep water canal. In the experimental work conducted by the University of Michigan group, the times for successful treatment of a hog carcass with radiation from mixed fission products varied from 320 min for the hind quarter to 1185 min for the fore quarter. Since a 24-hr period is usually allowed in meat packing plants from the time a hog is eviscerated and then cooled prior to cutting, this elapsed time was used profitably by the irradiation treatment of the carcass.

To test the biological effectiveness of this process in the destruction of encysted trichina larvae, feeding experiments were conducted with rats. Five thousand larvae which had been recovered from a sample of irradiated meat were fed by stomach tube to a group of rats. The control rats received 5000 larvae from non-irradiated meat. Examination of the small intestine of these rats was made 6 days after feeding. Thirty days later the muscles of these animals were examined for evidence of encysted larvae. The results of

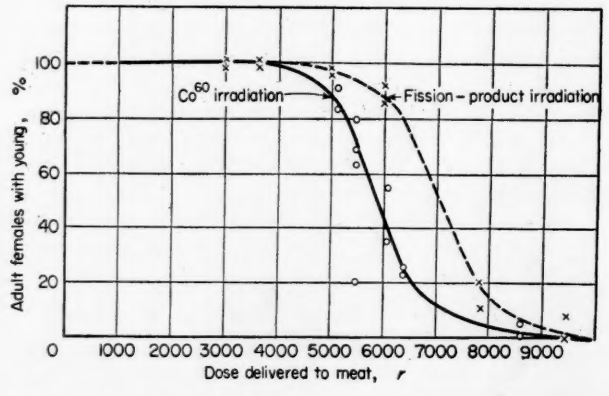


FIG. 6. Sterilization of trichinae in pork by $Co^{60}$ and waste fission-product irradiation. Dose measured by ferrous sulfate dosimeter (Lusteroid). Female adults recovered from rat gut 6 days after rat was fed irradiated larvae. (Reproduced from a paper by GOMBERG H. J. *et al.*[21])

these experiments are shown in Figs. 5 and 6. Using the $Co^{60}$ source, it was found that a dose of 11,000 rads was required for complete sterilization of the female trichinae, whereas with the mixed fission product source complete sterilization of females appeared to be accomplished at 9300 rads.

The irradiation of the pork has the added advantage of greater refrigerator shelf life as determined by taste panel tests. With irradiation doses up to 93,000 rads, no differences in flavor were detected by a taste panel when comparing the treated product with unirradiated pork.

This group of investigators has calculated the cost of this operation based on a plant that has an output of 2000 hog carcasses per day. These calculated costs included interest on investment with allowance for 5-yr amortization period. The irradiation cost per carcass would be approximately 2·3 mills or 0·23 cent/lb for a 135-lb finished carcass (excluding lard).

The importance of this process as a public health measure is readily appreciated. Despite statements to the contrary, a significant segment of the population consumes raw pork, causing trichinosis to be a public health problem. Convincing evidence for this statement has been provided by pathologists who have reported encysted trichinae in 16 per cent post-mortem examinations.

## REDUCTION IN ALLERGENICITY OF PROTEINS BY MEANS OF IONIZING RADIATION

*Gross anaphylaxis experiments*

In our laboratory, studies have been completed dealing with the determinations of the serological activity of $\gamma$-irradiated milk proteins which were treated at 0·475, 2·79, 5·58 and 9·30 Mrads. The technique for sensitization and challenge of guinea pigs has been described elsewhere.[22] The mean lethal shocking dose ($LD_{50}$) was determined for control (unirradiated) and $\gamma$-irradiated milk samples. These data are presented in Table 12.

Milk samples that had been irradiated with a $Co^{60}$ source were received from Dr. Goldblith (Massachusetts Institute of Technology). These irradiated milk samples were quite acceptable from the standpoint of flavor, odor and colloidal properties. In this process, the off-flavor and off-odor compounds formed during irradiation were removed by concurrent distillation. Only one irradiated sample which was treated at 9·30 Mrads was tested. The mean lethal shocking dose ($LD_{50}$ on guinea pigs) was determined for this sample and compared with the values reported for the $\gamma$-irradiated samples as shown in Table 12.

Presumably, the $LD_{50}$ values for 9·30 Mrad $\gamma$-irradiated milk (reported in Table 11) and those for the MIT sample should be comparable if they received the same radiation energies. Conditions may be variable enough, both in type of milk used and the dosimetry, for these two sources to account for some differences. However, no completely acceptable explanation for these differences is available at this time.

*Schultz–Dale experiments*

Another technique used for checking antigenicity of proteins is to challenge protein sensitized uterine strips. Uteri from 89 passively sensitized guinea pigs was challenged in the Schultz–Dale apparatus. Some of these animals were used for screening tests, but most were used to complete the comparative study of the effect of $\gamma$-irradiation versus thermal processing on alteration of milk proteins.

Raw skim milk in glass, screw-cap vials was packed and sealed in No. 2 metal cans, frozen, and then shipped with dry ice to the Gamma Irradiation Facility at Idaho Falls, Idaho. Four batches of milk were irradiated at dosages of 0·465, 2·79, 5·58 and 9·30 Mrads respectively. Immediately after irradiation, the cans were repacked in dry ice, returned to our laboratory, and stored in a freezer.

The heat processing was done by placing a glass, screw-cap vial of raw skim milk into a short length of close-fitting steel pipe

TABLE 12. Mean lethal shocking dose ($LD_{50}$) for milk sensitized guinea pigs challenged with $\gamma$-irradiated, concurrent radiation distilled* and raw skim milk

| Series | Radiation dose $10^6$ rads | Number of guinea pigs | $LD_{50}$ $\mu$g Protein N per dose | Approximate ratio Control vs. Irradiated |
|---|---|---|---|---|
| 1 | 0 | 66 | 36 ± 7 | 1 : 0·8 |
| | 0·475 | 66 | 29 ± 4 | |
| 2 | 0 | 77 | 32 ± 4 | 1 : 2 |
| | 2·790 | 87 | 64 ± 11 | |
| 3† | 0 | 64 | 69 ± 16 | 1 : 9 |
| | 5·58 | 50 | 602 ± 270 | |
| 4 | 0 | 60 | 37 ± 4 | 1 : 31 |
| | 9·30 | 55 | 1157 ± 422 | |
| | | *Values for concurrent radiation distilled milk* | | |
| 5 | 0 | 68 | 25 ± 4 | 1 : 5 |
| | 9·30 | 71 | 131 ± 23 | |

* Whole milk irradiated with $Co^{60}$ source and concurrently distilled for removal of off-odors and flavors at Department of Food Technology, Massachusetts Institute of Technology, Cambridge, Mass., U.S.A.

† Based on stock of antisera of lower antibody titer.

which was then sealed at both ends with threaded plugs. This sealed pipe containing the milk sample was immersed for 35 min in a thermoregulated, hot oil bath adjusted to the processing temperature. Only eight heat treated samples were prepared in this series—one at each 10° temperature increment from 60°C through 130°C. After cooling under cold tap water, the processed samples were stored in a freezer along with untreated control milk samples.

Four guinea pigs, yielding eight pairs of observations, were used for each of the twelve experimental samples. Each of these samples was simultaneously compared to the control milk response on a matching half-horn of uterine tissue. After the initial anaphylactic contractions for each were measured, a cross-challenge was made to determine whether the control milk would elicit a contraction in the strip which was previously desensitized by the experimental milk. By employing this technique, two different sets of comparative data were obtained—the initial response and the cross-challenge response. Small initial contractions and large cross-challenge contractions indicate the greatest degree of protein alteration.

The results of this study are summarized in the treatment response curves shown in Figs. 7 and 8. These are freehand curves drawn from the means of the eight pairs of observations collected from each experimental sample. Because of the few points and the high standard deviation for several of the means, these curves are quite limited and close comparisons may be of questionable value; but with this in mind, the following thermal treatment equivalents for $\gamma$-radiation dosages were extracted from the curves. The 9·30 Mrad sample had an antigenic response approximately equal to 89°C for 35 min; however, the cross-challenge response equivalent was only 78°C. For the 5·58 Mrad dosage, both the initial and cross-challenge response values were found to be near 77°C. The 2·79 and 0·465 Mrad samples were not sufficiently altered to make such comparisons.

With the gross anaphylaxis techniques being conducted at our laboratory, the antigenic response for a 90°C sample similarly processed was found to lie between the values for the 5·58 and 9·30 Mrad samples. This might indicate that the Schultz–Dale technique, as used in this study, tends to underestimate the protein alteration caused

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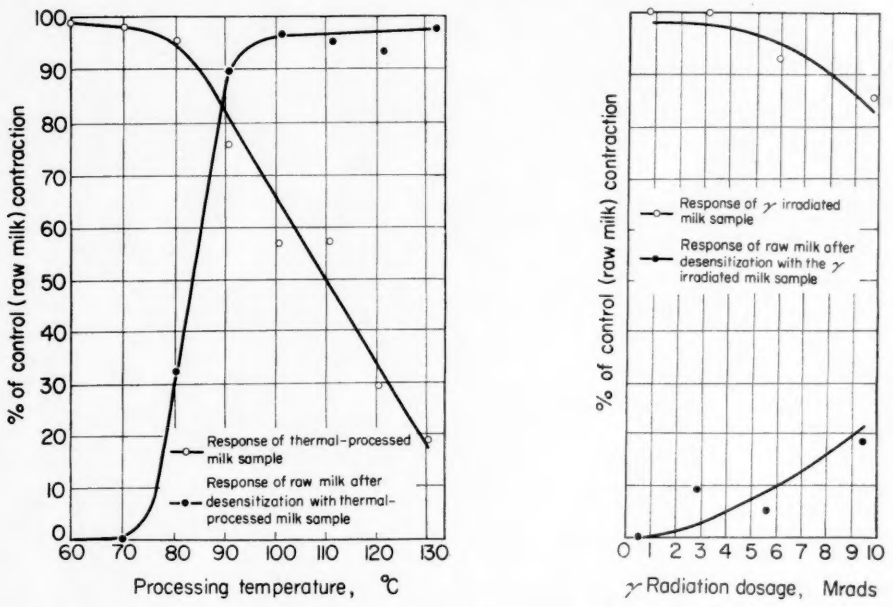
% of control (raw milk) contraction
Response of thermal-processed milk sample
Response of raw milk after desensitization with thermal-processed milk sample
Processing temperature, °C
Response of γ irradiated milk sample
Response of raw milk after desensitization with the γ irradiated milk sample
γ Radiation dosage, Mrads

FIG. 7. Antigenic response of guinea pig uterine tissue when challenged with milk proteins processed by heat and $\gamma$-radiation.
(a) Thermal processed (35 min) at various temperatures. (b) $\gamma$-irradiated at various dose levels.

by $\gamma$-irradiation; or that the gross anaphylaxis technique used in our other study underestimates the protein alteration caused by thermal processing.

Both heat processing and irradiation treatment of proteins are effective in altering proteins. The application of ionizing radiation offers a new approach to making available for clinical use food proteins with reduced allergenic properties.

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# Some Notes on the Wholesomeness of Irradiated Potatoes Fed to Pigs

T. HORNE and J. R. HICKMAN
Technological Irradiation Group, Isotope Division,
Wantage Radiation Laboratory, Wantage, Berks

(1) Pigs have been fed control and irradiated potatoes from 11 weeks of age till ready for slaughter for bacon production. The irradiation dose was 10,000 rads.

(2) The weight gains and food conversions for the pigs are given for the period of the experiment. No gross differences were observed.

(3) Blood samples taken at intervals showed no detectable differences in haemoglobin, urea, non-protein nitrogen, P.C.V. or plasma cholesterol.

(4) The bacon quality of the pigs was assessed in terms of the commercial value and no differences were observed.

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THE advantages of irradiation for the prevention of sprouting in potatoes are well known. There are large quantities of poor quality potatoes wasted each year. These potatoes are not used extensively as pig food in this country, chiefly because they are available for only limited periods each year. If, however, the storage period for potatoes were increased ensuring adequate supplies throughout the year, it is believed that they would be more extensively used as animal feedingstuffs. It would appear, therefore, that the preservation of potatoes by irradiation might be of economic importance in this respect.

The object of the studies, the first part of which is being recounted here, was to feed potatoes to pigs being fattened for the bacon market. The general health and the bacon quality of pigs fed irradiated and control potatoes were compared. The control potatoes used were those available at the appropriate season of the year, where no special storage precautions had been taken, apart from protection against frost. Where the control potatoes had sprouted, the sprouts were removed prior to cooking in order to minimize the risk of toxic effects attributable to the sprouts.

## METHODS

Two groups of six pigs were used. Each group contained one female and five males. The experiment was started when all the pigs were 11 weeks old. Each group of pigs was housed collectively, and the diets were fed on a communal basis. The food consumption was recorded each day and the animals were weighed once weekly.

The diets used were based on the supply of $2\frac{1}{2}$ lb of basal meal to each pig each day. Potatoes (variety "Majestic") were then supplemented *ad libitum*. The basal meal consisted of 70 per cent barley meal, 20 per cent wheatings and 10 per cent white fish meal. The potatoes were cooked by steaming prior to feeding. The potatoes fed to the pigs receiving the irradiated diet were irradiated with $\gamma$-rays from $Co^{60}$ at a dose level of 10,000 rads $\pm 10$ per cent. The potatoes were irradiated at least 2 weeks before feeding, to allow the reducing sugar content to fall to its pre-irradiation level.

At intervals, throughout the experiment and at slaughter, blood samples were taken from the vena cava for biochemical determinations.

When the pigs reached bacon weight, they were sent to the bacon factory for slaughter and grading. A gross morphological examination was made immediately after slaughter.

## RESULTS

TABLE 1. The average weight (lb) and the total live weight gain (lb) for pigs

| Age (weeks) | Irradiated | | Control | |
|---|---|---|---|---|
| | Average weight (lb) | Total gain (lb) | Average weight (lb) | Total gain (lb) |
| 11 | 45 | — | 46 | — |
| 12 | 51 | 32 | 52 | 39 |
| 13 | 58 | 43 | 61 | 52 |
| 14 | 67 | 53 | 71 | 59 |
| 15 | 73 | 37 | 75 | 29 |
| 16 | 82 | 58 | 86 | 61 |
| 17 | 90 | 49 | 95 | 55 |
| 18 | 101 | 66 | 106 | 65 |
| 19 | 112 | 62 | 113 | 44 |
| 20 | 123 | 66 | 127 | 86 |
| 21 | 132 | 57 | 136 | 53 |
| 22 | 143 | 65 | 148 | 71 |
| 23 | 156 | 77 | 157 | 53 |
| 24 | 170 | 85 | 169 | 71 |
| 25 | 179 | 50 | 175 | 37 |
| 26 | 193 | 84 | 188 | 77 |
| 27 | 205 | 72 | 200 | 77 |

*Live weight gains*

The live weight gains are shown in Table 1.

*Food conversion values*

The food conversion values were calculated on a basis of 1 lb meal being equivalent to 4 lb potatoes. The food conversion is:

$$\frac{\text{food consumed (lb) for 6 pigs}}{\text{live weight gain (lb) for 6 pigs}}$$

TABLE 2. Food conversion values

| Age (weeks) | Irradiated | Control |
|---|---|---|
| | Food conversion value | Food conversion value |
| 12 | 4·1 | 3·4 |
| 13 | 3·8 | 3·2 |
| 14 | 3·2 | 3·1 |
| 15 | 5·1 | 6·6 |
| 16 | 3·5 | 3·3 |
| 17 | 4·3 | 3·8 |
| 18 | 3·4 | 3·4 |
| 19 | 3·8 | 5·4 |
| 20 | 3·6 | 2·9 |
| 21 | 4·8 | 5·1 |
| 22 | 4·5 | 4·1 |
| 23 | 4·1 | 6·0 |
| 24 | 3·8 | 3·9 |
| 25 | 6·3 | 7·9 |
| 26 | 3·9 | 4·3 |
| 27 | 4·8 | 4·5 |

*Blood analyses*

Blood samples were taken for analysis during the first week of feeding and subsequently at approximately six weekly intervals and again at slaughter. Blood samples were collected in heparinized tubes. Haemoglobin, blood urea, non-protein nitrogen, P.C.V., Plasma S.G. and plasma cholesterol were determined. At no time were any significant differences noted between the irradiated and control groups.

*Bacon quality*

The pigs were sent to a commercial bacon factory for slaughter. The values of the pig carcasses are shown in Table 3.

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TABLE 3. The carcass value of pigs at slaughter

| Pig No | Date of slaughter | Weight Score | Weight lb | Price per score s. d. | Total price £ s. d. | Group |
|---|---|---|---|---|---|---|
| 5684 | 1/7/58 | 8 | 8 | 46 5 | 19 9 11 | C |
| 5673 | 1/7/58 | 8 | 3 | 42 11 | 17 9 9 | C |
| 5638 | 8/7/58 | 8 | 9 | 43 11 | 18 11 1 | C |
| 5674 | 8/7/58 | 8 | 2 | 46 5 | 18 16 0 | C |
| 5667 | 8/7/58 | 7 | 15 | 46 5 | 17 19 9 | C |
| 5675 | 22/7/58 | 7 | 10 | 45 5 | 18 10 8 | C |
| 5655 | 1/7/58 | 8 | 9 | 45 5 | 19 3 9 | I |
| 5668 | 1/7/58 | 8 | 5 | 42 11 | 17 14 1 | I |
| 5637 | 8/7/58 | 8 | 1 | 43 11 | 17 13 6 | I |
| 5651 | 8/7/58 | 7 | 19 | 46 5 | 18 9 0 | I |
| 5672 | 8/7/58 | 8 | 6 | 46 5 | 19 5 3 | I |
| 5679 | 15/7/58 | 7 | 18 | 46 5 | 18 6 8 | I |

C = Pigs receiving control diet.
I = Pigs receiving irradiated diet.

## DISCUSSION

The object of the experimental design was to rear pigs under conditions similar to those which might be used in commerce. This was achieved by housing the animals in groups and feeding a basal diet of "concentrates" with potatoes fed to appetite. The animals were kept under farm conditions and hence no special precautions were taken with regard to temperature control. This probably accounts for the very low weight gains and high food conversions in week 15 when the ambient temperature was very low. In spite of precautions to ensure that sprouts were not fed, it was suspected that some batches of potatoes in week 24 were not efficiently desprouted. The pigs refused to eat much on these occasions and hence the low weight gain.

The quality of bacon can be judged from the price received, although it must be borne in mind that the price per score varied throughout the period of slaughter. The price per score rose slightly between 1 July 1958 and 8 July 1958 and then remained fairly steady during the rest of the period of slaughter. It is of interest to note that the pigs receiving irradiated diets fetched £110. 12. 3 against £110. 17. 2 for the control pigs.

The figures for weight gains and food conversions have not been given after the twenty-seventh week, because subsequently pigs were sent to slaughter and the pigs remaining were observed to pine very badly and this was reflected in the weight gains.

*Acknowledgements*—The authors wish to express their thanks to the Director and staff of the Agricultural Research Council's Establishment at Compton for providing the pigs and for their help in housing and providing for the animals throughout this work.

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# Some Notes on Wholesomeness Trials at Wantage

J. R. HICKMAN
Technological Irradiation Group, Isotope Division, Wantage Radiation Laboratory, Wantage, Berks

THE object of this paper is not to present a mass of results on feeding trials with irradiated foods but rather to examine the approach to such feeding trials. Radiation damage to foods is likely to be slight. From the studies already reported by Dr. Kraybill, it must be concluded that no obvious changes will occur in foods from a safety point of view, although nutritional damage might be expected to some extent with proteins, fats and certain vitamins. The importance of nutritional damage must be assessed in each individual case against a background of the significance of that nutrient damaged from the food in question in the normal human diet.

Especially in low-dose procedures, such as procedures used for sprout inhibition in potatoes and procedures for insect control, the changes produced by irradiation are likely to be minimal in foodstuffs. Any changes which do occur are unlikely to be of great importance, but on the other hand, such changes must be known in order that their effects may be assessed. The chief concern, therefore, is to make the tests for nutritional damage and chronic toxicity as critical as possible. The methods for the assessment of nutritional quality are well known and will not be further considered here.

One of the main problems in assessing chronic toxicity is concerned with reducing the variance of response in a group of animals. Large variability in the results means that small changes will be masked and remain undiscovered. The design of multigeneration tests should be such that the differences detected are as small as possible and this will depend on the attention paid to the following factors:

(1) Number of animals per group.
(2) Reduction of all variables other than experimental treatments to a minimum.
(3) Design such that the effects of unavoidable sources of error can be estimated.

*Animals*

The rat is particularly well suited to this type of work. However, all too frequently rats used in biological research are unknown quantities. Our rats are obtained from our own stock colony kept under the same conditions as our experimental animals. In this way, we have strict control over animals used in experimental work and have data available of degree of inbreeding, normal variance of growth, fertility, disease and survival. These data serve as an additional control to our experimental work although, of course, not obviating the need for normal controls. Further, from these data regarding our stock animals, we hope to be able to select our breeding animals to improve the strain for our own particular type of work.

*Numbers*

The accuracy of an estimated mean varies with the square of the number of animals in each group. The standard error is reduced by a factor of approximately 0·7 by doubling the number of animals involved. Examination of the effect of numbers on standard error shows a rapid reduction in the standard error to the 20–30 animals per group level. After this the net reduction in error per animal decreases appreciably with increasing numbers. A compromise must be achieved



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between the number of animals used on one hand and the facilities available to make maximal use of those animals on the other hand. We have decided to use groups of 30 in this respect and since we are measuring factors which vary between the sexes (e.g. growth) we use 30 animals of each sex. In addition, we include a further 20 animals of each sex in each group for detailed histopathological examinations. Thus animals may be taken for histopathological examination throughout their life cycle without interfering with the main group being used for growth studies etc. In this way we hope to get a more complete histopathological picture and not simply results from animals which have finished their useful experimental life in other respects.

*Reduction of other variables*

Initial weights of experimental animals should be uniform; on the other hand, animals for multigeneration tests should be selected at random from a large number of litters and distributed amongst the groups at random. At Wantage we use the largest possible number of litters for selection, and discard any unusually small or large rats (from comparison with our stock records). The litters to be used are then selected at random and littermates are distributed amongst the groups at random. The corresponding members of any two subgroups are then littermates, and the effects of litters can be eliminated in the statistical appraisal. It has been found that elimination of litter differences can result in an improvement in accuracy equivalent to doubling the numbers of animals used.

*Growth*

Interpretation of growth is somewhat difficult. ZUCKER *et al.* [1–3] formulated a method of plotting growth graphically to give a straight line response. In practice, the curves obtained often, in our experience, show departures from linearity but these can be of value in the assessment of abnormalities in growth. Some causes of departure from the straight line are briefly discussed below.

(a) *Disease.* Animals suffering from disease frequently show departures from the linear plot; this shows very obviously in the Zucker plot. Departures from the linear plot often show the presence of subclinical disease and the animals can be sacrificed before gross symptoms appear. This means that the animals can be autopsied before gross morphological changes (or autolysis) mask interesting results. Any animals with subclinical infections must be sacrificed since they are unsuitable for further study. Using groups of animals with subclinical disease introduces unknown variables into the results. The only exception we make to this rule is where animals have mild respiratory diseases. Unfortunately, our colony is not free from mild respiratory disorders but these seldom lead to severe reactions in rats less than 1 year old. From records, the incidence of respiratory disease is known and any increase over this level would be viewed with alarm since, in an experimental group, this might suggest stress factors due to experimental treatments.

(b) *Realimentation.* Realimentation most frequently occurs at the immediate post weaning stage and lasts till the animals are up to 12 weeks old. This is caused by inadequacies on the part of the mother during the lactation period. The more adequate the diet the more rapid the rate of realimentation. This fact might prove of value in the evaluation of toxicity if an optimal basal diet be used. This point is at the moment being investigated.

(c) *Inherent weaning weight.* Variance in the weaning weights of rats is due to at least two causes, and since the weaning weights are used as a criterion of efficiency of lactation, some thought must be given to this problem. Variation in weaning weights can be attributed to (i) inherent differences in rat size and (ii) differences due to inefficiencies in lactation by the mothers. If Zucker's hypothesis is correct, then the linear plot, extrapolated to weaning time should give the inherent weaning weight. The difference between this figure and the actual weaning weight ought to give some measure of the efficiency of lactation by the mother. Work to investigate this possibility is in hand.

*Conclusion*

The need for a more critical approach to the appraisal of chronic toxicity cannot be denied. Some of the factors measured in multigeneration tests have been briefly discussed. Such tests must be further extended to include routine tests of specific functions, especially the liver, kidney, C.N.S. and blood.

Adequate control of experiments must be maintained and strictly this demands the use of both negative and positive controls. Suitable means for positive control have largely to be developed and at the present time, regretfully, it seems that positive controls will be used only where, from preliminary tests, specific damage is suspected.

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# Discussion

Mr. GREENWOOD (U.K.): What clinical studies of protein metabolism were made with human subjects; for example, was the amino acid content of the urine measured?

Dr. KRAYBILL (U.S.A.): No, the biochemical studies with human subjects were not extensive. When compared with a 2-year study on the rat, 15-day experiments with man covered only a very limited fraction of his total life span.

Mr. BORNAS (Spain): I should like further information on the numbers of animals which should be used on feeding trials of this nature for the results to be of statistical significance.

Dr. KRAYBILL (U.S.A.): In the early experiments with rats fed irradiated food diets, we proposed according to U.S. Food and Drug Administration recommendations that each test group have 50 animals (25 male and 25 female rats). In the long term feeding experiments using dogs and monkeys, it was recommended that 4 animals (2 males and 2 females) be employed for each group. As our national program progressed which involved using large numbers of animals and tonnages of foods, statisticians suggested the use of a factorial design employing one control group and eight test groups (total of nine groups) in which two irradiated foods could be tested simultaneously. To test two foods by the original plan of 50 rats per group, this would require 150 rats per food, 50 rats in the control group, 50 rats in the "X" irradiated food (3 Mrads) group and 50 rats in the "2X" irradiated food (6 Mrads) group. Hence, for testing two foods, this would require 300 rats.

Using the factorial design of nine groups with intercorrelations and comparisons on two foods, this would necessitate 20 animals per group with three times this number in the control group (60) or a total of 220 rats. The obvious advantage, therefore, in the testing of two foods is the saving in numbers of animals and quantities of food to be fed. However, this system suffers the marked disadvantage in not providing sufficient numbers of animals for histopathological investigations and enough numbers of animals with which to draw more accurate conclusions with respect to incidence of spontaneous tumor formation, if any. For the latter reasons, we would recommend the simple system of using 50 rats per group and 150 rats for the complete test and only testing one food at a time, with no intercomparisons between two foods at different radiation levels.

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# 5. NATIONAL FOOD LEGISLATION AFFECTING THE USE OF IRRADIATED FOODS AND PROGRAMMES AND PLANS FOR RESEARCH ON THE USE OF IONIZING RADIATIONS FOR FOOD PRESERVATIONS

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# A Survey of the United Kingdom Legislation Affecting the Use of Irradiated Food

W. M. SHORTT
Ministry of Agriculture, Fisheries and Food, Scientific Adviser's Division,
Great Westminster House, Horseferry Road, London S.W.1

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THE treatment of food with ionizing radiations is a recent development and it is not surprising therefore that specific legislation governing the use of these radiations in respect of food, or conditions applying to the treated food itself, are non-existent. It should not be assumed that the consuming public is not protected in some measure by existing legislation. The definitions of "food" in the Food and Drugs Act, and certain sections of the Act, are couched in general terms, and are capable of being construed as applying directly to food treated with ionizing radiation in common with all other food. Section 2 of the Food and Drugs Act of 1955 states that it shall be an offence if a person sells to the prejudice of the purchaser any food which is not of the nature, or not of the substance, or not of the quality demanded.

It is likely that the last named term, "quality", would be construed as including not only the dietary value of the food in question, but also its nutritional value. Should the effect of exposure to radiation result in any alteration of the nature or substance or quality of the food of such a magnitude that prejudice to a consumer could be established, a Court would be likely to hold that an offence had been committed. Owing to the nature of the irradiation process, and the effective controls which are certain to be imposed on dosage of radiation, it is unlikely that, except in extreme cases, either the "nature" or the "substance" of the irradiated food would be called into question. Changes in "quality" with its subsidiary aspects, the dietary or the nutritional value are more pregnant with possibility of action, since small alterations of composition, as for example, the decrease in vitamin content, or alteration of amino acid structure or content, as well as changes of texture, digestibility, flavour and the like could be embraced by the flexible term "quality".

In any proceedings under Section 2 the defences set out in Section 3 may be invoked. Where a food has been subjected to "process or treatment, other than food thereby rendered injurious to health, it shall be a defence . . . to prove that the operation in question was not carried out fraudulently, and that the article was sold having attached thereto a notice of adequate size distinctly and legibly printed and conspicuously visible, stating explicitly the nature of the operation, or was sold in a wrapper or container displaying such a notice". If therefore the irradiated food were sold adequately labelled a defence would be established against any action under Section 2 of the Act.

Control of statements appearing on a label which either falsely describes the food, or which is misleading as to nature, substance or quality, is effected by Section 6 of the Food and Drugs Act. A new offence would be created under this section if it could be shown for example that a food labelled in conformity with Section 3 had not been irradiated, or had been irradiated insufficiently; in other words, an attempt to forestall action under one section, could lead to the creation of an offence under another section of the Act.

Section 1 of the Food and Drugs Act deals

with offences in connexion with the preparation and sale of injurious foods, and states "no person shall add any substance to food ... or subject food to any other process or treatment so as to render the food injurious to health, with intent that the food shall be sold for human consumption in that state". This section poses a problem the proof of which is difficult in all cases, except those involving admixture of the food with a frank poison. The only means available for demonstrating whether irradiated food is, or is not injurious to health would seem to be some form of biological feeding tests. The limitations of this test are now well known, and the examination of any single variety of irradiated food would involve large numbers of appropriate experimental animals whose diet would accommodate the food under examination.

The test would have to be carried on for at least three generations, and framed in such a way that the final results were amenable to a rigid statistical analysis. Modern biological testing calls for the employment of several species of animals, and severe restriction of choice of animal may be imposed by the nature of the food under investigation, especially if, as is desirable, the irradiated food is to form the major part, if not the whole, of the animal's diet. All the resources of analytical chemistry, biochemistry, pathology, histopathology or other allied sciences would have to be employed during the testing period, since the effect to be demonstrated is likely to be small. Finally there is the unenviable task of relating any effect produced in the experimental animal to the probable effect in human beings, or alternatively, correlating the absence of any effect with a probable absence of effect.

One novel feature of the 1955 Act is a direction (Section 1 (5)) to be observed when assessing whether an article of food is injurious to health

> "... regard shall be had not only to the probable effect of that article on the health of a person consuming it, but also to the probable cumulative effect of articles of substantially the same composition on the health of a person consuming such articles in ordinary quantities".

This direction could have the effect of magnifying considerably the importance of any finding observed in the experimental animal, and would no doubt lead to some admirable legal disquisitions on the meaning of "probable cumulative effect".

Since irradiation is a method of preserving food it is pertinent to enquire whether the current Preservatives Regulations have any bearing. These Regulations issued in 1925 contain a definition of preservative which reads as follows "any substance which is capable of inhibiting, retarding or arresting the process of fermentation, acidification or other decomposition of food, or of masking the evidence of putrefaction". It is an offence to manufacture for sale, or sell any article of food, or to import for sale any article of food, which contains an added preservative (as defined above). (The exceptions to this general thesis are listed in the Regulations and do not concern us here.) Notwithstanding the comprehensive nature of the definition of "preservative", it is doubtful whether it could encompass within its scope the exposure of food to a beam of ionizing irradiation as constituting the addition of a preservative. But, having regard to the various theories advanced to explain the mechanisms involved during irradiation, it is not impossible that irradiated food may contain highly reactive radicals, peroxides, hydroperoxides or other chemical entities which, in their own right, would fall within the definition of preservative. If this is so, it is a nice point whether the induction of such bodies within the food by mere subjection to ionizing rays could be held in law to constitute "an addition".

Provided the presence of a chemical entity, falling within the definition of "preservative", could be demonstrated in an irradiated food, and was not there prior to irradiation, it seems likely that a Court would hold an infringement of the regulations had occurred. A pertinent point is the complete absence of any quantitative measure from the definition of "preservative"; it is of no avail to argue that the substance

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produced within the food is in such minute quantities that it could exert no preservative action whatsoever. The definition merely states "a substance capable . . ." and the capability may be demonstrated without regard to concentration.

The food before irradiation will invariably be contained in a wrapper or a container; one or other of the newer plastic films seems to be favoured as suitable material particularly for the envelope type of package. The Food Hygiene Regulations 1955, in addition to defining "container" and "contamination" stipulated that food shall not be wrapped in any wrapper so as to cause contamination of the food. The term "contamination" includes "contamination by odour" and experimental work to date seems to indicate that prevention of the development of foreign odours in a food during, or after, subjection to a dose of ionizing radiation is a difficulty. The Food Hygiene Regulations are concerned primarily with the contribution effected by the packaging material, but as the irradiation of prepackaged food seems to be the goal, it would be difficult to disentangle the effect due to the container, were an odour foreign to the food found after irradiation.

An authorized officer of a council may, at any reasonable time, examine any food intended for human consumption with a view to ascertaining whether the food is unfit for human consumption. Under Section 8 of the Food and Drugs Act 1955 it is an offence to sell, offer, expose or have in possession any food intended for human consumption but unfit for that purpose. Irradiated food would be subject, as with all other food, to this examination; and, if a Justice of the Peace agree, to the seizure and condemnation of the unfit food. Whether the food is or is not unfit for human consumption, is a matter of the opinion held by the examining officer; there is no obligation to require further examination by, say, the Public Analyst.

A similar situation arises in respect of imported food; the Public Health (Imported Food) Regulations 1937 prohibits the import into England and Wales of any article of food which has been examined by a competent authority (usually the Medical Officer of Health) and found to be unfit for human consumption. Once more, the opinion of the examining officer only is the deciding factor but before the condemnation is entered the order of a Justice must be secured. Admittedly the real value of these proceedings lies in dealing with foods which are grossly changed from the normal condition, usually foods which have decomposed or have become putrid, but these circumstances could arise for some types of irradiated food, particularly when coming from abroad.

In the event of the irradiation of food being permitted, presumably Food and Drug Authorities would be required to examine samples to see whether conformity to the various sections of the Food and Drugs Act had been observed. In the light of present knowledge any examination would have to be based on chemical and physical tests. Bacteriological tests would have limited value since the dosage of radiation applied to the food may have been designed to secure some effect other than that of complete sterility. It is unlikely that any Food and Drugs Authority would undertake to prove that a sample of irradiated food was injurious to health unless some test became available other than the biological feeding test. The limitations of the test itself are well recognized, the time element is unfavourable to Food and Drugs procedure, and the expense would be high. This can only imply that before irradiation can be regarded as an established practice, proof must be available that the submission of a given food to a stated optimal dose of radiation, results in no harmful effects on the food itself; and that the food, when eaten, is free from any suspicion that it may be injurious to health. This could mean that the general provisions of the Food and Drugs Act may need to be supplemented by a special regulation.

The Ministers of Health and of Agriculture, Fisheries and Food may, so far as appears to them to be necessary or expedient in the interests of public health, or otherwise for the protection of the public, make regulations for

(1) requiring, prohibiting or regulating the addition of any specified substance or any substance of any specified class . . . or the use of any such substance as an ingredient in the preparation of food and generally for regulating the composition of such food

(2) for requiring, prohibiting or regulating the use of any process or treatment in the preparation of any food intended for sale for human consumption . . .

The powers given to the Ministers could be exercised in a number of ways; one way would be to prohibit the subjection of food to ionizing radiations, if they were convinced that such prohibition were expedient in the interests of public health, or were needed for the protection of the public. If, however, irradiation of food were to be permitted as a treatment, the scope of any regulation would probably be determined by the actual amount of information and scientific data available on the treatment of food. It seems inescapable that the irradiation conditions and process would have to be linked quantitatively with the food or foods in the regulations, and that new foods or new processes would be admitted to the regulations as and when the requisite evidence was made available.

The work of the Food and Drugs Authorities would then be simplified to determining whether the conditions imposed by the regulations had been observed. The control of the heat treatment of milk (Pasteurized and Sterilized Milk Regulations 1949) affords a good example of the treatment of a close parallel problem, albeit relating to only one food—milk. The conditions of the treatment as regards temperature and time are specified, processes other than the basic ones may be used "with the approval of the Minister", records are required to be kept, and automatic control of the process can be stipulated. It is fortunate that in milk there is an "internal indicator" which is relied on to furnish evidence of the process to which the milk has been subjected—in the case of pasteurized milk the phosphatase and methylene blue tests, and the turbidity test for sterilized milk.

It is not beyond the realms of probability that discriminative tests parallel to these may be discovered for some if not all foods intended to be treated by radiations. In the absence of any such test one solution would be to create artificially in the food, or the container, the conditions leading to the recognition of the efficiency of the irradiation process, such as the destruction (or creation) of an easily recognizable system. Whether such an expedient would be adopted is doubtful, nevertheless the Ministers have the power as stated above, to require the addition of a specified substance to a food.

The rapidity of growth of the application of ionizing radiation to food is such that ignoring its presence cannot be maintained much longer. Recognition has already occurred in the United States where the Act recently passed includes within the definition of "food additive" ionizing radiation. "Food additive" is defined as " . . . including any substance intended for use in producing, manufacturing, packing, processing, preparing, treating, packaging, transporting or holding food and including any source of radiation intended for any such use." The time cannot be longer delayed when similar recognition must be accorded by all countries now exploring the new sources of energy made available through modern discoveries.

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# United Kingdom Regulations Covering Safety in the Use of Radiation Sources

S. SMITH

H.M. Chemical Inspector of Factories, H.M. Factory Inspectorate,
19 St. James' Square, London S.W.1

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APART from the general provisions of the Factories Acts, 1937 and 1948, there are no legal requirements in criminal law in the U.K. to protect workers concerned with the use of ionizing radiations in the irradiation of foodstuffs and similar processes. Even these "general provisions" are non-specific, in that they are worded in such a way that *any* hazard can be dealt with, be it due to radiations, or poisons, or even dangerous plant or buildings.

But a draft Code of Regulations, the Factories (Ionizing Radiations) Special Regulations, has been published, as a first step towards a statutory final Code; and comments on the draft provisions have been received from Industry and other interested folk. The Code will deal with the use of sealed sources of radiations in factories, building sites, etc.; and, among other things, will apply to the irradiation of foodstuffs, in so far as the process entails risk to the workers doing the irradiation, but not in so far as the process might or might not contaminate in some way the products, and so cause danger to the consumer.

As it stands, the draft mentions irradiation processes specifically in its Part IV, which reads as follows:

## PRECAUTIONS RELATING TO CERTAIN PROCESSES

*Application of Part* IV *of these Regulations*

28. This Part of these Regulations applies only to: the use of ionizing radiations for radiography; the testing of X-ray or other apparatus or film; and the irradiation of materials for the purpose of sterilization or of inducing chemical, physical or biological changes of state.

*Requirements as to sealed sources*

29.—(1) Radioactive substances intended to produce mainly $\gamma$-rays shall be sealed sources with wall thickness sufficient to allow no more than one per cent of the primary $\beta$-rays to escape.

(2) A serial number or other identifying symbol shall be on or attached to every sealed source to distinguish it from all other sealed sources under the control of the occupier.

(3) When not in use or in transit or being tested sealed sources shall be stored under lock and key in a place that is reserved exclusively for the purpose and is adequately shielded.

(4) Where more than one sealed source is stored each shall be kept in a separate compartment which shall enable adequate protection to be afforded to all persons.

(5) Where work involving removal of a sealed source from its protective receptacle is being done the sealed source shall be moved from or returned to its protective receptacle only by the use of a handling rod, remote controls, or an automatic method, so as to ensure adequate protection to all persons concerned in these manipulations.

*Provision of enclosure for using ionizing radiations*

30.—(1) Subject to the provisions of Regulation 31 hereof, ionizing radiations shall be used only within an enclosure set apart for the purpose and providing under all operating conditions adequate shielding against direct and scattered radiation for all persons outside the enclosure.

(2) Whilst any sealed source within the enclosure is exposed effective arrangements shall be made to exclude from the enclosure all persons other than the radiology staff who shall, whilst ensuring adequate protection for themselves, enter or remain in the enclosure for the minimum time necessary to make essential adjustments to the apparatus within.

(3) Effective interlocks shall be provided and maintained to prevent any person from entering the enclosure whilst any X-ray apparatus within it is in operation.

(4) Means of exit from the enclosure shall be so constructed that any person accidentally shut inside can leave the enclosure without delay.

(5) Where X-rays are used in the enclosure, the X-ray control panels shall be situated outside it; and safe means shall be provided within the enclosure for interrupting quickly the electrical supply that enables the X-ray apparatus to be energized. Where a sealed source is used inside the enclosure, means shall be provided to enable any person accidentally shut in to control the mechanism whereby the sealed source is removed from and returned to its place of storage.

(6) Observation windows fitted into the enclosure shall be of lead glass or other suitable material so as to afford adequate shielding.

(7) Sealed sources for the irradiation of materials for the purpose of sterilization or of inducing chemical, physical or biological changes of state shall be stored within the enclosure.

(8) All materials and articles being treated by irradiation shall be brought into the beam of radiation or moved during such irradiation by the use of conveyors or other mechanisms operated from outside the enclosure.

*Radiography where provision of an enclosure is impracticable*

31. In the radiography of materials by means of X-rays or by means of ionizing radiations from a sealed source, where the provision of an enclosure in accordance with paragraph (1) of Regulation 30 hereof is impracticable, the radiography shall, as far as practicable, be isolated from other work, and the operator of an X-ray apparatus shall be adequately protected from direct and scattered X-radiation by a cabinet or shields or other means and other persons shall be adequately protected either by exclusion from a suitable marked area round the X-ray apparatus or sealed source and the article being examined or by other means:

Provided that where a sealed source is in use, members of the radiology staff may, whilst ensuring adequate protection for themselves, enter or remain in such marked area for the minimum time necessary to make essential adjustments to the apparatus within.

*Gamma-ray radiography*

32. In the $\gamma$-ray radiography of materials, the radiographic set-up shall be completed before the sealed source is exposed.

*Warning signals and notices*

33.—(1) Lights or audible signals, or both, shall be provided for the purpose of warning persons in the vicinity of the control panel, high voltage generating equipment and X-ray tube, where these are used, and persons in the vicinity of, or accidentally shut inside, the enclosure provided in accordance with paragraph (1) of Regulation 30 hereof. The said lights and audible signals shall be operated immediately before the exposure of the sealed source; or where the ionizing radiations are X-rays, they shall be arranged to operate automatically at a stage prior to the energizing of the high voltage generator.

(2) A separate warning light shall be exhibited at all times during the excitation of the X-ray tube or during the exposure of a sealed source as the case may be.

(3) Where ionizing radiations are used

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otherwise than in accordance with Regulation 30 hereof, legible warning notices capable of being read easily by persons in the vicinity shall also be displayed during the periods referred to in the preceding paragraphs of this Regulation.

These provisions are self-explanatory, except for a few of the terms that are used, e.g. "radioactive substance", "adequate protection", and" adequate shielding", which have special meanings, and are defined in Regulation 2.

"*Adequate protection*" means, briefly, that the worker must not get a higher dose than is recommended by the I.C.R.P.; and any means, including limitation of duration of exposure, may be employed to this end.

"*Adequate shielding*" has a similar meaning, except that it is assumed that, in the worst case, a worker will be near the relevant plant for the whole of his working life; so no question of limiting the duration of exposure arises.

Although the I.C.R.P.'s recommendations are easy to understand and are well known, they are not phrased in a form that can be transposed directly into Regulations: for example, they refer to the rem as their unit, which introduces the concept of relative biological efficiency. For this reason, and many others, the I.C.R.P.'s recommendations have had to be rephrased in precise legal form in a rather complicated Schedule, which is reproduced below; and these two definitions (of "protection" and "shielding") have had to be linked with it.

## SCHEDULE Regulation 2(2)*

### Permitted Radiation Doses Referred to in the Definition of the Expression "Adequate Protection" in Regulation 2(2)

*General*

1.—(1) For the purposes of this Schedule and the Table thereto, no account shall be taken of any ionizing radiations that do not arise from the processes.

(2) Where the occupier is able to show that a person not employed in the processes:

(a) is tested for personal exposure to ionizing radiations in accordance with Regulation 11 hereof; and

(b) is subject to medical supervision and examination as required by Regulation 22 hereof,

that person may be regarded as being employed in the processes.

(3) Any weekly dose specified in this Schedule or the Table thereto for any person may be exceeded in any week if the total of the radiation doses received by that person in that week and in the twelve weeks immediately preceding does not exceed ten times the weekly dose so specified so, however, that where the weekly dose is so exceeded the total of the radiation doses for that period of thirteen weeks shall not exceed ten times the weekly dose.

*Maximum permissible weekly doses for persons employed in the processes*

2. Subject to the provisions of sub-paragraph (3) of paragraph 1 and of paragraph 4 of this Schedule, where the ionizing radiations consist mainly of X-, $\gamma$-, or $\beta$-rays, the maximum permissible weekly dose for persons employed in the processes shall be 100 mrads. In the case of other ionizing radiations, the maximum permissible weekly dose for those persons shall be 10 mrads.

*Maximum permissible weekly doses for persons not employed in the processes*

3. Subject to the provisions of sub-paragraph (3) of paragraph 1 and of paragraph 4 of this Schedule, where the ionizing radiations consist mainly of X-, $\gamma$-, or $\beta$-rays, the maximum permissible weekly dose for persons not employed in the processes shall be 30 mrads. In the case of other ionizing radiation, the maximum permissible weekly dose for those persons shall be 3 mrads.

*Maximum permissible doses in certain factories*

4.—(1) Where in relation to any factory the occupier is able to show that the

* Since this talk was delivered the I.C.R.P. has modified its standards—the standards on which the schedule in this paper was based. No doubt the Final Code will take this change into consideration.

conditions specified in the following Table are observed, the provisions of that Table shall apply, instead of the provisions of paragraph 2 or paragraph 3 of this Schedule, with respect to the adequate protection of persons in that factory.

(2) For the purposes of the said Table, where in any week the types of ionizing radiations to which a person is exposed are not confined to those specified in any one serial number in the Table, the maximum permissible dose for that person in that week shall be in accordance with the weekly dose in serial number 1 of the Table. The dose which that person receives in that week shall be deemed to be the sum of the doses which that person receives from each type of ionizing radiation specified in the Table and doses due to neutrons, α-particles or protons shall be multiplied by ten before they are added to the doses received from other ionizing radiations.

(3) For the purposes of the said Table, where in any decade any person employed in the processes is exposed to ionizing radiations of types specified in both serials number 1 and 5 of the Table, the decade dose shall be in accordance with serial number 1. For the purposes of this paragraph, doses due to neutrons, α-particles or protons shall be multiplied by ten before they are added to doses received from ionizing radiations of

TABLE 1. This table is subject to the provisions of sub-paragraph (3) of paragraph 1 and of sub-paragraphs (2) and (3) of paragraph 4 of this schedule

| Serial no. | Conditions | | |
|---|---|---|---|
| | Type of ionizing radiations | For persons employed in the processes | For persons not employed in the processes |
| 1 | X- or γ-radiation with a half-value layer of 1 mm or more of water, other than the ionizing radiations that are referred to in Serial no. 3 of this Table. | (i) The maximum weekly dose shall be not more than 0·3 rad. (ii) No person shall receive more than 50 rad in any decade. | The maximum weekly dose shall be not more than 0·3 rad. |
| 2 | X- or γ-radiation with a half-value layer less than 1 mm of water. | The maximum weekly dose at the skin shall be not more than 1·5 rad. | The maximum weekly dose at the skin shall be not more than 0·15 rad. |
| 3 | X- or γ-radiation with a half-value layer of 1 mm or more of water, that is restricted mainly to the hands, forearms, feet, ankles, head (excluding the eyes) and neck. | (i) The maximum weekly dose at these parts of the body shall be not more than 1·5 rad. (ii) In so far as the ionizing radiations affect parts of the body other than those mentioned in the first column of this Serial, the conditions of Serial 1 of this Table apply. | (i) The maximum weekly dose at these parts of the body shall be not more than 0·15 rad. (ii) In so far as the ionizing radiations affect parts of the body other than those mentioned in the first column of this Serial, the conditions of Serial 1 of this Table apply. |
| 4 | β-rays | (i) The maximum weekly dose in the lenses of the eyes shall be not more than 0·3 rad. (ii) The maximum weekly dose at the skin shall be not more than 1·5 rad. | (i) The maximum weekly dose in the lenses of the eyes shall be not more than 0·03 rad. (ii) The maximum weekly dose at the skin shall be not more than 0·15 rad. |
| 5 | Neutrons, α-particles or protons | (i) The maximum weekly dose shall be not more than 0·3 rad. (ii) No person shall receive more than 5 rad in any decade. | The maximum weekly dose shall be not more than 0·003 rad. |

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the types specified in serial number 1, and doses due to ionizing radiations other than those specified in serials number 1 and 5 shall be left out of account.

"*Radioactive Substance*" has not hitherto been defined satisfactorily in British Law (nor elsewhere, so far as can be ascertained by cursory enquiry). Previous definitions say that any emitter of ionizing radiations is a radioactive substance. But, on this definition, our very bodies are radioactive substances, in that they emit radiations from the minute amounts of potassium-40 that they contain. We had, therefore, to find a specific activity that was so high that ubiquitous weak emitters would be excluded; yet sufficiently low to include all the genuinely radioactive substances that could be hazardous if inhaled or ingested under foreseeable conditions. The figure of 0·002 $\mu$c/g was chosen.

This figure was derived by dividing the 40 hr non-occupational m.p.c. for plutonium (the most toxic element) in air ($6 \times 10^{-7}$ $\mu$c/m$^3$), by 0·3 mg/m$^3$ which latter figure was felt to be a reasonable average concentration of "process dust" in a factory. Although dust concentrations of more than 10 mg/m$^3$ occur in some very dusty factories, not all of this is "process dust"; concentrations of 0·1 to 1 mg/m$^3$ are by far the most common ones for "process dust", and 0·3 is roughly the geometric mean of 0·1 and 1.

With a specific activity of $2 \times 10^{-3}$ $\mu$c/g (parent), and assuming 1 meV of $\beta$- or $\gamma$-radiation per disintegration, the surface dose rate would be about 2 mrads per hour. Thus a person could spend his entire working life fairly near to a mass of this material without exceeding the maximum permissible dose. Hence it seems unnecessary to limit the definition by an over-riding level of radiation intensity at the surface of the substance.

Although Part IV of the draft Code deals specifically with the irradiation of foodstuffs, and certain other named processes, the previous parts of the draft Code also apply in that they apply to all processes to which the Code applies. These earlier parts are fairly straightforward, and introduce no new concept: provision is made for regular medical examinations, for personal monitoring, for monitoring of shielding, for prohibiting employment before the age of 18 years, and for the appointment of a competent person (not necessarily of the standard of a "health physicist", in many cases).

There are certain administrative provisions, such as the notifying of intention to use sealed sources for the first time in a factory, etc. and some general requirements as to the construction, storage, and use of sealed sources.

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# Individual Country Statements on National Food Legislation Affecting the Use of Irradiated Foods

## INTRODUCTION

In the section which follows, individual statements from eleven separate countries are included. From these it will be seen that, while in the case of some countries the sale of irradiated food is forbidden by law, in several other countries there is no food legislation which refers specifically to the production and sale of irradiated food, though existing food legislation could, where necessary, be applied to such foods. This is also the case for some other countries in Europe (e.g. Belgium, Italy and Portugal) for which separate country statements on food legislation do not appear in the present publication.

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## FRANCE*

Food irradiation, which from the legal point of view is considered as a chemical treatment, is at present forbidden by French legislation. The ban extends to the sale of foods so processed, irrespective of their origin.

Before the processing and sale of such foods can be made permissible, the agreement of the National Health Council and of the Academy of Medicine will have to be obtained.

Such permission could be granted only on condition that:

(a) There exists a compelling necessity to use such methods of processing and their efficiency in attaining the desired objectives is established with certainty.
(b) Evidence is supplied that irradiation does not alter the nutritive value of foods, nor affect undesirably their organoleptic properties. Possibility of controlling that the food was really irradiated.
(c) Thirdly, and above all, it has been demonstrated beyond any doubt that irradiation cannot have any dangerous effects on consumer health, both short and long term.

Finally, should the use of ionizing radiations be authorized, the doses applied would have to be kept to the minimum necessary and mention that the food was irradiated should appear on the label.

If the desired objective of harmonizing national regulations is to be attained, it appears desirable even now to provide for international co-ordination of research and experimentation between research workers and health specialists, on the basis of a protocol to be worked out in advance.

* Prepared by C. Gross, Paris.

## FINLAND

At present there exists in Finland no special legislation covering the use of irradiated foods. If necessary, however, the production and the sale of irradiated foods can be prohibited under the Food Act, issued July 3, 1941, and under the Food Statute of November 21, 1952 (No. 408). Clause 9 of the Food Statute reads as follows:

"The production and sale of foods injurious to health is forbidden. Food shall be regarded as being injurious to health: (1) if it can cause poisoning or other illness or if, owing to spoilage, contamination or faulty manufacture, or some such reason, it must be considered unfit or unsuitable for human consumption".

The following legislation is in force in Finland as regards irradiation protection: the Act of Irradiation Protection, April 26, 1957 (No. 174), the Statute on Protection against Irradiation, and the Decision of the Ministry of the Interior referring to Irradiation Protection, issued March 15, 1958. This legislation covers protection against dangers from irradiation.

## FEDERAL REPUBLIC OF GERMANY

The Food Act of 5 July 1927 (Reichsgesetzblatt I, p. 134) in force up to now does not contain any special stipulations on the use of ionizing or ultra-violet rays in the food industry. Only quite generally—i.e. also with regard to the use of rays—the following things are prohibited:

(1) To obtain, produce, process, pack, store or transport foodstuffs for others in such a way as to render their consumption injurious to human health (paragraph 3, section 1a) and

(2) To offer, store for sale, expose for sale, sell or circulate in any other way foodstuffs under misleading names, descriptions or presentations (paragraph 4, section 3).

The new Food Bill, which was passed by the Bundestag on 7 November 1958 and which will presumably come into force towards the end of the year,* provides the following stipulations for the use of ionizing and ultra-violet rays.

*Paragraph 4c, section 1*

Foodstuffs may only be treated with ionizing or ultra-violet rays in so far as this is expressly permitted. Such a process may be permitted if it is compatible with the protection of the consumer, either generally or for single foodstuffs or for special purposes.

As far as the marking is concerned, the new law provides for the following:

*Paragraph 4c, section 2*

Foodstuffs which have been irradiated must be marked. The method of marking is provided for in the Orders in Council in section 1. Exemptions from this obligation to mark can be permitted in such Orders in Council, if the kind and doses of the exposure do not give rise to concern and the consumer cannot be misled in his expectations, if the labelling is omitted.

In the official explanatory preamble of this law it is mentioned that the exposure of foodstuffs to ionizing and ultra-violet rays is licensed, because the effect of these rays can at the moment not yet be fully overlooked.

*Exemptions*

According to paragraph 5a of the new law, exemption from the obligation to mark foodstuffs which have been irradiated by ionizing and ultra-violet rays may be permitted in so far as this is compatible with the protection of the consumer.

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* The new Food Law was published on 21 December 1958 and is now in force.

## IRELAND

Ireland has not enacted legislation dealing specifically with the use of irradiated foods. Should the question arise of statutory control of the use of ionizing radiations for food preservation, it is possible that control could be exercised at present under the provisions of existing food legislation. A summary of this legislation is set out below:

(1) Under the Sale of Food and Drugs Acts, 1875–1936, it is an offence to mix, colour, stain or powder any article of food with any ingredient or material so as to render the article injurious to health with intent to sell the article in that state or to sell to the prejudice of the purchaser any article of food which is not of the nature, substance and *quality* of the article demanded. It is a matter for the Court before whom prosecutions are brought for sale of a sample of food alleged to be non-genuine to decide whether an offence has been committed.

(2) The Food Hygiene Regulations, 1950 and 1952, prohibit the sale for human consumption of food which is diseased, contaminated or otherwise unfit for human consumption, and require that adequate precautions shall be taken to prevent the contamination of food intended for sale for human consumption at all stages of its manufacture and distribution, including the maintenance of hygienic conditions in food premises and the observance of certain hygienic precautions by food handlers. The Regulations provide for the seizure of unfit food, and for its destruction; and also for the registration of food premises.

(3) The Public Health (Saorstát Éireann) (Preservatives, etc., in Food) Regulations, 1928, and the Public Health (Saorstát Éireann) (Preservatives, etc., in Food) (Amendment) Regulations, 1943, prohibit the addition of preservatives to food (with limited exceptions in certain foods).

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## THE NETHERLANDS

There exists for the time being no special decree affecting the use of irradiated foods in the Netherlands. If necessary, however, the sale of such foods can be prohibited immediately under the General Municipal Food Inspection Decree, as laid down in article 6 of the Commodity Act, whereas importation of such foods can be prevented under article 16 of the Commodity Act. Another possibility which is feasible on short term is an extension of article 3 of the General Decree of the Commodity Act.

Article 3 of the General Municipal Food Inspection Decree forbids the selling of foods which are or might be hazardous or harmful to health, if used corresponding to their destination.

The decision whether certain products are hazardous or harmful is left to the discretion of the Directors of the Food Inspection Services.

As there do not exist tolerances for radioactivity in foods and regulations with respect to changes in food composition and properties of irradiated foods so far, decisions on these matters must be reached in the courts.

Article 16 of the Commodity Act is concerned with the importation of foods and relates to the protection of public health and makes it possible to enact a decree to forbid or to permit the importation conditionally. Furthermore, in the latter case any importation must take place along detailed custom houses. If necessary the importation of irradiated foods can be stopped immediately by issuing such a decree.

In article 3 of the General Decree of the Commodity Act are laid down requirements in respect of the machines or equipment etc. used in food manufacturing and of their effects on foods. An extension of this General Decree of the Commodity Act is feasible.

As has already been said a special decree

does not at present exist; however, a general Atom-act is being prepared, which will make it possible to issue decrees covering all hazards which may arise from the application of ionizing radiations for food processing and food preservation.

## POLAND

There is no separate legislation concerned with food irradiation. In this field there is applicable only one injunction of the Ministerial Council of 23 May 1957 for security and hygiene, when working with ionizing radiations; it is given in the *Journal of Laws of the Polish Republic* No 34, June 27, 1957.

This enactment divides all laboratories using a free source of ionizing radiations into three groups depending on the amount of isotopes used. The maximal permissible doses of radiation are stated. They are given as 0,3 r per week and 0,05 r per day for X- or $\gamma$-rays (quantum energy below 3 MeV); 0,3 r per week for $\beta$-radiation (molecule energy below 3 MeV); 0,3 rad per week for X-, $\gamma$- or $\beta$-radiation (quantum or molecule energy above 3 MeV).

The maximal permissible concentrations of radioactive substances in potable water and air, when constantly exposed to hazards, are given in detailed tables.

The frequency of determining the amounts of radiation received by workers is stated according to the group in which the laboratory is classified.

Detailed regulations are given for X- and $\gamma$-radiations in industrial establishments. Daily dose received by a worker must not exceed 0,05 r. With a free source of radiation the radiation level must not exceed 1 mr/hr.

Precautionary measures—both general and individual—in isotope laboratories are given at length (gloves and special clothing, goggles, shower-baths, air-filters; Geiger counters in rooms, covers). Appendices are given, e.g. protective shieldings (thickness in millimetres with different materials) around sources of X-rays, $\gamma$- and $\beta$-radiations.

## SWEDEN

According to the general regulations in paragraph 7, clause 1 of the Swedish Food Regulations, issued 21 December 1951, no substance may be offered for sale as food or transferred to another person for sale as food, if, as a result of deterioration, contamination, faulty preparation, or for any other reason, it may be assumed to be dangerous to consume or otherwise unfit for human consumption.

Whether this regulation is applicable to irradiated foods depends on whether it may reasonably be assumed that generally or in a specific case these foods are dangerous to consume or otherwise unfit for human consumption. The burden of proof is on the authorities.

In certain circumstances, however, the regulations in paragraph 7, clause 3, will be applicable to irradiated foods, though primarily they have biological warfare in view. The clause reads: "When the kingdom is at war or in danger of war, or if otherwise required by extraordinary circumstances, the Crown may order that a given substance or a substance from a given place, if such substance may be expected to cause disease or transmit infection, or if other special reasons exist, shall not be offered for sale as food nor be transferred to another person for use as food. If such order has been given, the substance shall be subject to the same provisions as apply to a substance the sale of which is prohibited in accordance with clause 1 above.

If the kingdom is at war, an order as said in the first paragraph may be given by an authority prescribed by the Crown.

If a Health Committee of County Council considers that reason exists to issue an order in accordance with the first part of this clause, the Committee or County Council shall give immediate notice thereof to the Crown or to such other authority as the Crown may appoint to issue such order."

## SWITZERLAND

Foodstuffs which have been subjected to some physical processing (for instance ionizing, ultra-violet or ultrasonic waves) may only be put on the market after the process has been approved by the Federal Health Office (FHO).

Approval will only be given if it can be proved that no harmful change takes place, nor any modification in the foodstuff which may be contrary to the regulations laid down in this order.

Periodical biological tests must be made in a Swiss institution recognized by the FHO, at the expense of the producer or vendor, to ensure that no substances with toxic or other undesirable effects are present and to check on the intensity of the treatment.

The process used must be clearly described on the package, or otherwise clearly shown if the goods are on sale without packaging.

## U.S.A.

Public Law 929, dated September 6, 1958, "an Act to protect the public health by amending the Federal Food, Drug, and Cosmetic Act to prohibit the use in food of additives which have not been adequately tested to establish their safety", defines in Section 201, paragraph S the word "additives", specifically mentioning irradiation: "and including any source of radiation intended for any such use". Section 402 (a) has had added the words "if it has been intentionally subjected to radiation . . ." It should be noted, of course, that older legislation, such as the Meat Inspection Act, which enables the inspectors of the Department of Agriculture to inspect and, if necessary, condemn, is still valid.

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# Individual Country Statements on Programmes and Plans for Research on the Use of Ionizing Radiations for Food Preservation

## BELGIUM

*Facilités disponibles pour le traitement des aliments*

Le Centre d'Etude de l'Energie Nucléaire à MOL est à la disposition des chercheurs.

*Les travaux en cours et envisagés*

Les principales études en cours sont les suivantes:

(a) Recherches de mutants progressifs sur Hordeum Sativum et sur Triticum vulgare. Par la Station de Recherches de l'Etat pour l'Amélioration des Plantes de Grande Culture, rue du Bordia, à Gembloux. *Directeur:* M. E. LAROSE. *Chefs de Travaux:* A. MOES et L. NOULARD.

(b) Recherches sur le métabolisme dans les fermentations de divers produits laitiers. Par la Station laitière de l'Etat, à Gembloux. *Directeur:* M. R. PIRAUX. *Chef de Travaux:* P. JAMOTTE. *Assistant:* R. LACROSSE.

(c) Recherches sur l'influence de l'irradiation des denrées par les neutrons sur les insectes.
Recherches sur l'influence des rayons-$\gamma$ sur les insectes par la Station d'Entomologie de l'Etat, à Gembloux (collaborant avec le Centre de Recherches Hautes Fréquences). *Directeur:* M. E. VAN DEN BRUEL. Chef de Travaux: D. BOLLAERTS.

(d) La Station de Chimie et de Physique agricoles de l'Etat, rue du Bordia, à Gembloux (*Directeur:* M. G. DEMORTIER) a chargé un de ses collaborateurs (M. RIGA) de se spécialiser dans l'utilisation des radiations. Celui-ci fait actuellement un stage de 1 an aux U.S.A.

(e) Le Professeur PEETERS de l'Ecole de Médecine vétérinaire de Gand utilise les radioisotopes dans ses recherches sur la physiologie de la lactation chez la vache.

## FRANCE*

1. *Les sources d'irradiation*

A part une source de 600 c de $Co^{60}$, qui est à Lyon, aucune autre source n'est réservée exclusivement à la stérilisation alimentaire; cependant, au Centre d'Etudes Nucléaires de Saclay, deux sources de 600 c de $Co^{60}$ ont déjà servi pour des essais. D'autre part, en 1959 sera mise en service à Saclay une casemate d'irradiation utilisant les éléments combustibles usés de la Pile EL3; une partie de cette casemate, conditionnée en température, sera spécialement réservée à la stérilisation alimentaire. En outre, cette même année, une source de 3000 c sera mise en fonctionnement dans une station située près de Lyon; cette source sera, plus tard, portée à 20,000, puis à 40,000 c. Enfin, deux générateurs d'électrons peuvent être utilisés en partie pour la stérilisation alimentaire: il s'agit d'un accélérateur de 4 MeV avec

* Préparé par G. MOCQUOT, Station Centrale de Recherches Laitières et de Technologie des Produits Animaux, Jouy-en-Josas (S. & O.), France.

une puissance de faisceau de 400 W (Ets. Massiot) et d'un générateur électrostatique Sames de 600 keV, donnant des courants d'électrons de quelques mA. Pour l'instant, les études de dosimétrie sont faites par le CEA, mais déjà certaines sociétés privées se chargent de telles études.

2. *Les travaux récents ou en cours*

Un certain nombre de travaux récents, effectués par des chercheurs français, intéressent notamment les études fondamentales de bactériologie. Ils sont susceptibles d'apporter des informations pour les recherches appliquées au traitement des denrées alimentaires par les radiations ionisantes. Ces travaux ont trait à l'action protectrice exercée chez les bactéries par diverses substances vis-à-vis des radiations ionisantes;[1,2] à l'efficacité biologique relative de divers rayonnements;[3] à "l'effet oxygène";[4] à l'influence de divers facteurs—et spécialement des facteurs nutritifs—sur la survie des micro-organismes irradiés;[5,6,7] enfin, aux propriétés biologiques et immunologiques particulières des bactéries stérilisées par les rayonnements ionisants.[8]

A la Sorbonne, le Professeur Haissinsky a étudié le mode d'action des radiations sur des systèmes physico-chimiques et ce travail est de grande importance pour la compréhension des phénomènes qui se passent avec les substances alimentaires.

D'autre part, des travaux de radiobiologie fondamentale concernant les modifications physico-chimiques subies par les protéines au cours de l'irradiation, sont effectués actuellement par le Service de Biologie du Centre d'Etudes Nucléaires de Saclay.

Sur le plan des applications, l'Ecole des Industries Agricoles et Alimentaires étudie, en liaison avec le Service de Biologie de Saclay, les modifications apportées par l'irradiation au métabolisme et aux activités enzymatiques des pommes de terre et des oignons au cours de leur conservation, ainsi que l'action de l'irradiation sur le pool des constituants solubles et sur les activités enzymatiques du blé et de la farine, en rapport avec les modifications de leur valeur boulangère.

Les recherches poursuivies au Laboratoire de l'Inspection Technique des Subsistances concernent:

(1) l'évolution des amino-acides et de la vitamine C dans les pommes de terre traitées par irradiation (en vue d'inhiber la germination);
(2) les modifications de composition des acides aminés des viandes irradiées.

Au Laboratoire de Biochimie et Physico-chimie des Céréales (Institut National de la Recherche Agronomique), on a abordé l'étude des modifications subies par les principaux constituants des grains et des farines de céréales au cours de l'irradiation: en liaison avec le Laboratoire de l'Ecole Française de Meunerie, on a examiné l'influence de la dose de radiations reçues par les farines de blé sur la valeur boulangère et les propriétés physico-chimiques des constituants plastiques ou dynamiques (plus particulièrement amidon, lipides, protéines insolubles).

Au Laboratoire du Froid à Bellevue, le Professeur Ulrich a étudié l'influence des radiations sur la respiration des fruits et des légumes. Cette recherche est en relation directe avec la radio-pasteurisation des tissus.

D'autres données, concernant l'aspect pratique des problèmes posés par l'irradiation des denrées en France, ont déjà été exposées,[9] en particulier la question de l'irradiation des pommes de terre[10] et l'influence des emballages.[11]

Ces études d'application ont été poursuivies par la Société Conservatome et ont permis la mise au point de certaines techniques d'irradiation, ainsi que la recherche de doses convenables pour chacun des produits étudiés.

3. *Les études envisagées par les différents laboratoires et services de recherches*

Parmi les études envisagées, celles qui doivent être considérées comme distinctes des études déjà en cours concernent les points suivants:

Au Service d'Hygiène Atomique et de Radiopathologie du Commissariat à l'Energie

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Atomique, recherches sur la toxicité éventuelle des denrées alimentaires.

Au Laboratoire de l'Inspection Technique des Subsistances:

(1) détermination d'un éventuel indice de peroxydes et des dérivés sulfurés ou carbonylés des denrées irradiées;
(2) essai d'identification des produits pouvant être considérés comme responsables des odeurs et saveurs anormales constatées dans les denrées traitées.

Au Laboratoire de Biochimie et Physicochimie des Céréales (INRA) les modifications produites, dans l'amidon et dérivés, par l'irradiation, en fonction de la durée d'exposition et de l'intensité du rayonnement, en présence ou en absence d'oxygène ou d'autres substances. On aura en vue l'obtention possible de produits présentant des propriétés et des domaines d'utilisation nouveaux.

On envisage également l'étude de l'action des radiations-$\gamma$ sur le déroulement normal de la réaction de Maillard et l'étude de l'influence d'une irradiation préalable des constituants glucides–protides, en milieu synthétique et dans les milieux naturels à base de céréales, sur leurs interactions.

Le rôle du degré de liberté de l'eau contenue dans les produits sur les altérations subies par leurs constituants au cours de l'irradiation sera particulièrement pris en considération.

A la Station de Technologie Végétale et à la Station Centrale de Recherches Laitières et de Technologie des Produits Animaux (INRA), recherches de bactériologie qui seront d'abord centrées sur l'action des radiations vis-à-vis des nombreuses souches déjà isolées de produits alimentaires, fermentés ou non.

Pour les bactéries utiles à l'élaboration des produits alimentaires—et, en particulier, celles que l'on utilise comme levains—on cherchera à préciser leur résistance aux radiations en fonction de l'état et de la composition du milieu sur lequel elles prolifèrent normalement.

Pour les micro-organismes, qui sont des agents d'altération, on recherchera, en outre, les moyens d'augmenter leur sensibilité. Cette dernière étape concernera, en particulier, les bactéries sporulées, aérobies ou anaérobies, qui sont susceptibles de jouer un rôle dans la conservation de diverses substances alimentaires. On cherchera dans quelles conditions on peut limiter ou arrêter leur développement en combinant l'irradiation à l'action de la chaleur, des antiseptiques, des antibiotiques et, d'une façon générale, des autres conditions du milieu (pH, teneur en eau).

On envisage aussi la recherche systématique et l'étude des germes résistant aux radiations, car ils semblent susceptibles de limiter l'efficacité des traitements.

Au Service de Biochimie et de Nutrition du CNRZ (INRA), on étudiera l'influence des radiations ionisantes sur l'oxydation des lipides et la valeur biologique des protéines dans les carcasses de poulets irradiés, ainsi que les substances responsables de l'odeur et de la saveur formées lors de l'irradiation de ces mêmes produits.

Au Laboratoire Municipal de Paris, les études envisagées concernent le contrôle des denrées irradiées. Ce contrôle est nécessaire, d'abord pour obtenir éventuellement l'autorisation de mettre en vente ces denrées et, ensuite, pour assurer la loyauté des transactions. Les recherches à cet égard pourraient être basées sur la modification des caractères analytiques intrinsèques des aliments sous l'effet de l'irradiation (apparition ou disparition de certaines substances) ou faire appel à des révélateurs, radiosensibles ou non.

Des recherches à ce sujet devront donc être entreprises.

Enfin, bien qu'il ne s'agisse plus d'applications concernant les pays d'Europe, l'ORSTOM s'intéresse à l'action des radiations sur les denrées végétales d'origine tropicale et, plus particulièrement, sur le plan entomologique, à la stérilisation et à l'éradication dans le cadre de l'ensilage des denrées alimentaires.

4. *Liste des organismes français* participant aux recherches sur la préservation des substances alimentaires par les radiations ionisantes

Centre de Recherches et d'Etudes du Génie Rural
Antony (Seine)

Commissariat à l'Energie Atomique (CEA)
Centre d'Etudes Nucléaires de Saclay (CEN)
—Service d'Hygiène Atomique et de Radiopathologie,
—Service de Biologie,
—Service d'Application des Radio-Eléments.
Boite Postale No. 2, Gif-sur-Yvette (S. & O.)

Conservatome
18, rue Seguin, Lyon (2°)

Ecole Nationale des Industries Agricoles et Alimentaires
9, rue de l'Arbalète, Paris (V°)

Institut National d'Hygiène
3, rue Léon-Bonnat, Paris (XVI°)
L'Institut National d'Hygiène, qui est chargé par la loi d'étudier et de promouvoir les recherches sur les facteurs alimentaires de la santé, a une section de nutrition organisée de façon à pouvoir assurer directement ou indirectement ce rôle.

Institut National de la Recherche Agronomique (INRA)
Centre National de Recherches Zootechniques
—Station Centrale de Recherches Laitières et de Technologie des Produits Animaux,
—Service de Biochimie et de Nutrition,
—Station de Recherches Avicoles
Jouy-en-Josas (S. & O.)
Laboratoire de Biochimie et Physico-chimie des Céréales
16, rue Nicolas Fortin, Paris (XIII°)
Station de Technologie Végétale
Route de Saint-Cyr, Versailles (S. & O.)

Laboratoire de l'Inspection Technique des Subsistances
6, boulevard des Invalides, Paris (VII°)

Laboratoire Municipal de Paris
39*bis*, rue de Dantzig, Paris (XV°)

Office de la Recherche Scientifique et Technique Outre-Mer (ORSTOM)
Service de Radiobiologie: Institut d'Enseignement et de Recherches Tropicales
80, route d'Aulnay, Bondy (Seine)

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## ITALY

Research in radiation preservation of food and its practical application are as yet at an early stage in Italy. However, plans and programmes of work sponsored by the public and private institutions which are directly interested in the field, are currently under way.

The National Council of Atomic Research is preparing the way for continuous work of its research centres. The reactor ISPRA I will be provided with high level gamma irradiation facility, which is the type of source most suitable for research with very high doses of irradiation such as those needed for food preservation. This radiation facility will be available in 1960, when it will be

possible to use spent fuel elements from the test reactor.

The National Council of Atomic Research, in co-operation with the Fruit and Vegetables Research Institute at Parma and the National Institute of Nutrition, has designed a research programme to study the possible changes in the organoleptic qualities and nutritional value of irradiated food, as well as the toxic compounds which may be produced by the irradiation process.

In connexion with the economics of food production and consumption in Italy, particular attention will be given to the products to be exported such as fruits, vegetables, canned tomatoes, as well as to the food to be imported, such as meat, eggs and fish.

Italy is also greatly interested in the application of ionizing radiations for the disinfestation of grain.

The current programme of work is supervised and encouraged by the following organizations: the Italian Ministry of Agriculture, the Italian Ministry of Health, the Ministry of Industry and Commerce, the National Council of Atomic Research, the National Research Council, the National Productivity Council and the Fruit and Vegetable Research Institute at Parma.

## POLAND

*1. Radiation facilities available for the treatment of food*

Until 1958 there were not in Poland any radiation facilities available for the treatment of food, and the newly installed atomic pile at Swierk near Warsaw does not offer any such possibilities in the near future either in the way of utilizing its waste radiation or in the production of radioactive isotopes.

At the Electrotechnical Institute (Industrial Radiological Laboratory) of the Warsaw Polytechnical School, there is a newly installed $Co^{60}$ source of about 720 c designed for the examination of metallic structures, which, to some extent, can be utilized also for the radiation treatment of food at the highest rate of 900 rads/hr.

There are, also, several rather small X-ray facilities, e.g. at the Chair of Physics of the Central College of Agriculture in Warsaw, which may be used for the studies of the effects of small doses of ionizing radiations on food, tissues and micro-organisms.

*2. Work in progress*

At the end of 1958 some investigation on the control of sprouting of potatoes and onions had been started and for the past 2–3 years research work on the action of X-rays on the enzymatic systems and on some tissues has been continued.

Research on the application of ionizing radiations to foodstuffs in doses appropriate either for radiopasteurization or for sterilization has not yet been started.

*3. Plans for future work*

It is generally acknowledged that large-scale and independent investigations on the food irradiation will only be possible after the installation of radiation facilities specially designed for the purpose. Until such facilities become available the work outlined below will be initiated (or continued) according to the existing radiation possibilities, due consideration being given to both the practical significance and theoretical value of the planned topics of investigation, viz:

(1) Investigating the practical possibility of increasing the commercial keeping qualities of prepared foods by treating with small doses of $\gamma$- or X-rays (20,000 to 100,000 rads).

(2) Investigating the possibility of improving the microbiological quality of rennet cheese milk by using radiation of 20,000 to 100,000 rads, principally for the purpose of destroying about 95 to 99 per cent of the *Coli* group of bacteria (the relatively high sensitivity of these micro-organisms having already been proved in earlier studies, e.g. by DUNN *et al. J. Appl. Phys.* **19,** 605 (1948)).

(3) Observations on the action of small doses of $\gamma$- and X-rays on the rennet coagulability of milk, on the physical and chemical

properties of the clot, and on the ripening of hard cheeses made therefrom.

(4) Conducting observations on the formation of permanent mutants of *Oospora lactis* and *Penicillium camemberti* moulds in reference to their vitaminogenic capabilities (thiamine, riboflavin and niacine).

(5) Investigating, at a later stage, formation of propionic bacteria mutants as concerns production of cobalamines and active vitamins $B_{12}$, under the influence of X- or $\gamma$-rays.

(6) Observations on retarding the sprouting in potatoes and onions by the small doses of X- or $\gamma$-rays (5000 to 20,000 rads), with special consideration to the commercial and dietetical value of treated products.

*4. Institutions engaged on research on irradiation of foods*

There is not a distinctly defined centre directly concerned with the radiation treatment of food. It has been decided, however, to concentrate this work within the existing Agricultural Isotope Laboratory at the Central College of Agriculture in Warsaw (Director—Prof. Dr. M. Górski).

Through the initiative of the Isotope Commission at the Agricultural Department of the Polish Academy of Sciences (P A N) and also by the sponsorship of the Ministry of Commerce, some actual research on the potatoes and onions has recently been started within the Soil Cultivation and Nutrition Institute (Vegetables Crops Laboratory in Skierniewice) with the utilization of the Radiation Laboratory in Warsaw.

In carrying out the programme of research the main responsibility will rest with the Chair of Agricultural Food Industries and the Chair of Physics of the Central College of Agriculture, in close co-operation with the Isotope Laboratory already mentioned, and also the Institute of Dairy Industry in Warsaw.

## SWEDEN*

The Royal Swedish Academy of Engineering Sciences has appointed a committee of seven members to follow the progress in irradiation preservation of food, to stimulate research on food irradiation in Sweden and to investigate the potentialities that irradiation preservation may offer for typical Swedish foods. The Swedish Food Irradiation Committee has as chairman Professor H. Lundin, and as secretary Dr. L.-E. Ericson, both of the Division of Food Chemistry, Royal Institute of Technology, Stockholm.

*1. Radiation facilities*

There is one van de Graaff accelerator in Gothenburg, one in Lund and two in Stockholm, but none of these machines is at present set up for electron acceleration. No substantial $\gamma$-source suitable for food irradiation research is yet available; the two largest in existence are the 200 c $Co^{60}$ at Bogesund (just outside Stockholm) and the 100 c $Co^{60}$ at Uppsala. Not until the end of 1959 will spent fuel elements be available from the 30.000 kW materials test reactor now being built at Studsvik (60 miles south of Stockholm), where also a 6 MeV van de Graaff accelerator and a 1000 c $Co^{60}$ source will be installed.

Research on food preservation by means of ionizing radiations is therefore at present hampered by the lack of radiation sources readily available to the food research institutions, and one of the primary tasks of the Committee will be to secure financial support for procuring such facilities.

*2. Work in progress*

Dr. Maire Jaarma, Institute for Organic Chemistry and Biochemistry, University of Stockholm, is continuing her studies on the influence of irradiation on the metabolism of potato tubers. She has found that irradiation causes an increase in the contents of sucrose and glucose but a decrease in the

* Prepared for the Swedish Food Irradiation Committee by H. Lundin and L.-E. Ericson.

concentration of fructose. Whereas a dose of 16.000 rads generally causes a definite and almost total inhibition of sprouting, the changes in the sugar contents of the tubers are reversible up to this dose level. The dose required to inhibit germination completely, depends on the variety, on the length of time after harvest and on the dose rate.

Dr. E. von Sydow, Head of the Swedish Institute for Food Preservation Research, Gothenburg, and his collaborators have recently begun work on the possibilities of controlling the degree of maturity in semi-preserved fish products. This is an item which is produced in considerable quantities in Scandinavia, both for export and for domestic consumption. Dr. von Sydow's group will also investigate the actual cause of the maturing processes, thereby making use of the relatively high sensitivity of micro-organisms to irradiation compared to enzymes. This work is being carried out in collaboration with Conservatome, Lyon, France, and the Atomic Energy Research Plant, Risø, Denmark.

Dr. L.-E. Ericson at the Division of Food Chemistry, Royal Institute of Technology, Stockholm, is studying the stability of vitamins, amino acids and enzymes when irradiated in the pure state and in foodstuffs. In collaboration with Conservatome investigations have begun on irradiation-induced changes in vitamin $B_{12}$. An attempt will be made to identify possible biologically active breakdown products of this vitamin.

Drs. L. Ehrenberg and G. von Ehrenstein at the Institute for Organic Chemistry and Biochemistry, University of Stockholm, and A. Ehrenberg at the Biochemical Department, Nobel Medical Institute, Stockholm, have carried out preliminary joint investigations of free radicals and peroxides in irradiated food. A method is being developed for the quantitative determination of free radicals and peroxides that can be used for studies of changes in the concentration of these compounds during storage, preparation and mastication of irradiated food. The chemically more stable end products are investigated, especially those from irradiated carbohydrates. Methods have been developed to demonstrate radiomimetic effects of free radicals, peroxides and their end products and to estimate the hazards for mutation and cancer in man. The formation of very reactive compounds in irradiated bread and certain model substances has already been demonstrated. Due to lack of grants these investigations have been discontinued for an unknown period of time.

Drs. P.-O. Kinell and O. Nilsson at the Institute of Physical Chemistry and the Institute of Physics, University of Uppsala, Uppsala, are also carrying out free radical investigations. They too are trying to work out a physical method for quantitative determination of free radical concentrations in irradiated material. Furthermore their studies are concerned with the identification of end products from irradiated proteins, peptides, amino acids, carbohydrates etc., and with the effects of various substances, e.g. free radical acceptors, on the course of the reactions induced by irradiation.

*3. Plans for future work*

There are no immediate plans for future work of another nature than that outlined under "Work in progress".

*4. Institutions engaged in research on irradiation of foods*

Biochemical Department, Nobel Medical Institute, Stockholm. Division of Food Chemistry, Royal Institute of Technology, Stockholm. Institute for Organic Chemistry and Biochemistry, University of Stockholm, Stockholm.

Institute of Physical Chemistry and Institute of Physics, University of Uppsala, Uppsala.

Swedish Institute for Food Preservation Research, Gothenburg.

## UNITED KINGDOM*

1. *Introduction*

The statement of plans in the United Kingdom for future work in this field is presented by the Ministry of Agriculture, Fisheries and Food (M.A.F.F.) speaking for the various bodies whose plans are discussed. It is an agreed statement but does not imply that M.A.F.F. is responsible in any way for the co-ordination of the work being carried out in any other organization but its own.

Means of irradiation on a large scale are now beginning to look promising from sources within the United Kingdom whether machines or radioactive isotopes are to be employed.

It is anticipated that supplies of $Cs^{137}$ up to a megacurie will be available by 1965. A large $Co^{60}$ source has already been sold to Australia at 10s. per curie. Cobalt-60 production may well become cheaper, in which case its price would compare favourably with the anticipated price of 1s. to 2s. per curie for caesium.

Provided that the problems associated with the food itself can be overcome, there should be an adequate supply of irradiation sources for treatment on a large scale.

2. *Ministry of Agriculture, Fisheries and Food plans*

The M.A.F.F. has for some time kept a continuous survey of the development of this technique by visits of its officers to the organizations conducting this work in the U.S.A. and the United Kingdom. A Food Irradiation Group, including in its membership U.K. workers in this field and representatives of the Armed Services, has been formed to keep the Services fully informed of current development work and of the potential future application of ionizing radiations to Service food supplies. It is anticipated that when medical clearance for the irradiated products has been given and they are becoming available on the civilian market, the Services will then assess the operational and financial advantages of using irradiated foods in comparison with those of conventional supplies.

The Ministry, however, is aware of the problems still awaiting solution and is constantly reviewing the possibility of initiating research on its own account; particularly into problems of a fundamental character.

Work being carried out at the M.A.F.F. Research Station and Experimental Factory at Aberdeen on the problem of oxidative rancidity, is concerned with the free radical reactions which initiate it and with their inhibition. Fundamentally there is common ground between this and some of the problems attendant on sterilization by ionizing radiations.

Most of the work has been devoted to the production of more efficient free radical acceptors and a small irradiation source (500 c $Co^{60}$) has been used to initiate oxidation on a small scale. In addition to the main line of work on unsaturated fatty acids and their esters, fresh milk has been tried as a substrate and the limited experience with this suggests that the off-flavour effects which arise with a dosage probably of the order of 500,000 rads may be avoided by the presence of the newly developed free radical acceptors. For other foods which can tolerate substantial treatments in any case (milk is notoriously sensitive) the additional dose made possible by the use of free radical acceptors may give that extra margin to permit a useful level of irradiation without rendering the product unpalatable.

This project will be pursued owing to its promise but the possibility of this Ministry taking a more active part in research into and practical application of the technique is now being reviewed afresh. The outcome will not be known for some months, but the time is now opportune in view of the expected availability of kilocurie sources being produced by the Atomic Energy Authorities.

3. *Department of Scientific and Industrial Research (D.S.I.R.)*

*Low Temperature Research Station (Cambridge).* At the Low Temperature Research Station

* Prepared by G. Wortley for the Ministry of Agriculture, Fisheries and Food, Scientific Adviser's Division.

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there are at present six staff employed on food irradiation work. Sources in use are two X-ray units (90 kV 80 mr), and a 2 MeV electron accelerator available nearby; a small $Co^{60}$ source is proposed.

Work in collaboration with the Ditton Laboratory of D.S.I.R. on potato sprouting has been terminated. Irradiation suppresses sprouting, but causes undesirable changes in quality; and a generally more satisfactory procedure has been developed at Ditton (treatment with nonyl alcohol vapour).

Small scale experiments on frozen eggs indicate that salmonellae can be eliminated with a dosage of 3–500,000 rads. Tests on a larger scale (about 1 ton of material) are being made in collaboration with the Central Public Health Laboratories, following which rat feeding trials will be carried out at the A.E.R.E. Laboratories, Wantage.

Experiments in radiation pasteurization of eviscerated poultry indicate that doses of 800,000 rads give about a five-fold extension of storage life at +1°C. About 130 ordinary consumers did not notice anything unusual in such carcasses when roasted, and a trained test panel does not usually distinguish between control and irradiated chicken, if tested soon after irradiation. The acceptability tests are to be continued.

Experiments are to be carried out to determine what effect additives to meat, in the form of curing salts and spices, have on the changes in quality resulting from irradiation, e.g. of sausages and bacon. Comparisons of irradiation of meat in oxygen with that in nitrogen have given confusing results; hence, instruments are being tested which might permit precise determination of the oxygen status of meat tissue. The nature of the various off-odours produced under different conditions is to be investigated with gas chromatography. Work on irradiation at different temperatures has suggested that whereas there is little difference in quality at temperatures above 0°C, below that, at a given dose, considerably less change results at lower temperatures. The greatest effect of temperature on irradiation changes occurs within the range 0 to −20°C and this range is to be more fully explored especially with regard to microbiological and chemical changes during storage.

Enzymic activity in irradiated food presents a problem: meat undergoes autolysis during storage. Studies of the inactivation of meat enzymes by radiation have been started, to elucidate the nature of these autolytic changes.

*Torry Research Station (Aberdeen).* At the Torry Research Station some experiments on the irradiation of fish have been made in collaboration with the Cambridge group. All the fish species examined showed adverse quality changes with doses greater than 1 Mrad, but cod and lemon sole showed comparatively small changes in quality after doses in the range 0·25–0·50 Mrad. Such doses give a useful increase in storage life at 0°C, particularly if combined with antibiotic treatment.

The effect of storage on the quality and the microbial flora has been studied. It is intended to continue investigations on the use of low doses of radiation in combination with antibiotic treatments, followed by storage under chilled conditions. Particular attention will be given to prepackaged fish fillets.

4. *Technological Irradiation Group, Atomic Energy Research Establishment*

In the Technological Irradiation Group of the Atomic Energy Research Establishment there are twenty staff working directly on foodstuffs and a further eight on the entomological aspects of grain storage in particular. The Group has one 4500 c and one 600 c $Co^{60}$ source, and 2kW 4 MeV linear accelerator and a fuel rod irradiation assembly using spent fuel rods from high flux reactors of the DIDO type. Future work is likely to be confined to $\gamma$-radiation as being the more useful and accurate research tool. By the summer of 1959 a pilot irradiation plant will be in operation using a 150,000 c $Co^{60}$ source and with a sterilizing (2 Mrad) capacity of 50,000 lb of material per week.

Of the work in progress, that on milk will shortly be suspended since pasteurization by

irradiation offers no advantage over traditional methods and sterilization doses give rise to marked off-flavour. Development, therefore, depends on the solution to this problem. What has been learned of the changes in certain protein fractions and altered digestibility of proteins can be applied to other foods.

Studies on small quantities of a wide variety of fruit and vegetables have been made. Doses of 250,000 rads on hard and soft fruits reduce mould wastage and increase storage life by 3–4 times. Other troubles which develop can be held in check by low temperature storage but further studies are needed. Changes of rate of maturation and textural changes which impose the upper dose limit will also be studied. Some limited work will continue on enzyme changes in potatoes rendered dormant by irradiation, and it is hoped to carry out a storage trial (50–100 tons) under commercial conditions next season. Since it seems unlikely that, for suppressing sprouting of potatoes, irradiation offers material advantages over chemical agents, no further work is planned.

The use of radiation in extending the storage life of prepackaged vegetables will be investigated; this will involve acquiring a suitable wrapping material. The fate of steroid compounds in irradiated foods will be followed in greater detail and identification and evaluation of the nutritional properties of irradiated products attempted.

Some fundamental work on the influence of radiation on alginic acid and its derivatives and their extraction from seaweed is under study. It is hoped to identify some of the volatile components of irradiation odours and flavours by gas chromatography.

In general emphasis will be on low doses. The possibilities for sterilization await success in preventing off-flavour and/or micro-organism sensitization. The use of mild heat treatment, low temperature storage and, with meat, adrenalin injection before slaughter to prevent enzyme activity is being investigated. Animal feeding trials which already include both whole foods and components will be extended to include specially treated foods, e.g. firstly, irradiated frozen whole egg and then soft fruits and a meat product will probably follow.

The emphasis on the entomological side will move from grain to higher cost items, e.g. dried fruit. Here too, to guard against reinfestation, emphasis will be on packaged goods. Although doses will be low, particular attention will be paid to the possible development of odours and flavours. Low doses give little control of micro-organisms and insect death may be delayed for up to 4 weeks so allowing further damage but the cost is lower and risk of off-flavours and odours very unlikely. The balance of advantages by use of high and low dosages will be evaluated for various products.

*Work at other centres*

The various academic centres in the United Kingdom are extending their work in this field now that radiation sources are becoming more readily available. A certain amount of fundamental work on the effects of ionizing radiations on micro-organisms is in progress in universities such as Cardiff, Manchester and London, while radiation chemistry is studied at several centres, e.g. Newcastle, Leeds, Edinburgh, Cardiff, London and Manchester.

## U.S.A.*

*1. Radiation facilities available for treatment of food*

The facilities listed below are described very briefly. Full addresses are given so that inquiries may be sent directly to the installation.

*Electron equipment*

(1) ARCO Western Service Irradiation Center, Applied Radiation Corporation, Walnut Creek, California
Linear accelerator 1·3 kW 2–10 MeV
19,000 lb/8 hr day

* Prepared by the Interdepartmental Committee on Radiation Preservation of Food.

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(2) W. F. and John Barnes Company, 301 South Water St., Rockford, Illinois
Linear accelerator 2·5 kW 2–10 MeV 78,000 lb/8 hr day
(3) General Electric Company, X-ray Department, 4855 Electric Avenue, Milwaukee 1, Wisconsin
Resonant transformer 2·4 kW 1–2 MeV 2000 lb/8 hr day
(4) High Voltage Engineering Corporation Radiation Facility, Burlington, Massachusetts
Van de Graaff accelerator 0·5 kW 0·75–2 MeV 2000 lb/8 hr day

*Other sources*

(1) High Level Gamma Irradiation Facility, Argonne National Laboratory, P.O. Box 299, Lemont, Illinois
(a) Mixed fission product 0·22–2·5 MeV 300 lb/8 hr day
and
(b) Fission product 0·22–2·5 MeV 300 lb/8 hr day
(2) Radiological Division, Dugway Proving Ground, Dugway, Utah
(a) Spent fuel rods 0·75 MeV 130 lb/8 hr day
and
(b) Cobalt-60 1·25 MeV 400 lb/8 hr day
(3) Phillips Petroleum Company, P.O. Box 1259, Idaho Falls, Idaho
Spent fuel elements 0·35–2·9 MeV 6400 lb/8 hr day
(4) Savannah River Plant, Atomic Energy Commission, Aiken, South Carolina
Spent fuel rods *ca.* 2 MeV 900 lb/8 hr day
(5) Radiation Laboratory, Stanford Research Institute, Menlo Park, California
Cobalt-60 1·16–1·33 MeV 14 lb/8 hr day
(6) Cook Electric Company, Inland Testing Laboratory, Morton Grove, Illinois
Cobalt-60 1·2 MeV

*2. Work in progress*

The American program for preservation of foodstuffs got under way after the end of World War II. The first article appeared early in 1947. References in technical and industrial literature now number by the thousands and have been conveniently assembled by the Quartermaster Food and Container Institute. These bibliographies are available as reprints from the Office of Technical Services, U.S. Department of Commerce under the general title "Ionizing Radiations: their production, effects, and utilization with special reference to food and packaging technology"—more than 1200 pages of text and over 500 pages of index—identified as PB 111635 ($6.75), PB 111636 ($8.00), PB 111637 ($7.25), PB 111636S ($11.00), PB 111637S ($4.25), PB 111636-S-2 ($4.75), and PB 111636-S-3 ($3.00).

The Department of Army, in co-operation with other interested governmental agencies investigated the possibilities inherent in the process. Since 1954 the Army, which has received the warm support of the Joint Committee on Atomic Energy of the Congress, has been engaged in an extensive program of research and development. To provide the co-ordination of the efforts of all governmental agencies and to assist in the transition from a government-sponsored program to the free economy the Interdepartmental Committee on Radiation Preservation of Food was established in May 1956.

Current work covers the broadest fields—sources, dosimetry, health physics, preservation of foods, standards of regulation and inspection, packaging, industrial education and preparation, construction of a pilot plant (the U.S. Army Ionizing Radiation Center at Sharpe General Depot near Stockton, California) at which production problems and costs will be worked out, government support and international relations. "Food Preservation by Radiation as of 1958", and "The Interdepartmental Radiation Preservation of Food Program: first report by the Interdepartmental Committee" (Feb. 1957), available as PB 131169 ($1.00), both add a great deal of detail to all aspects of the program.

*3. Plans for future work*

The outstanding immediate plan for the future is the USAIRC mentioned above. The planning for the operations, which are scheduled to start in the second half of 1960, is in the hands of Irradiated Products, Inc.,

a corporation set up expressly for this purpose and whose stockholders are four well-known companies—Armour & Co., Continental Can Co., Inc., Food Machinery and Chemical Corporation, and General Foods Corporation. This interest, coupled with the researches being carried on independently by various industries, gives adequate assurance of the present awareness of the food industry that a new process is in the making. In a country like the United States, where the canning and freezing and the distribution of foodstuffs is already very highly developed, it appears more probable that preservation or extension of shelf life by irradiation will be an evolutionary rather than a revolutionary development.

The following time table, which, it cannot be too strongly emphasized, is necessarily very tentative, indicates the goal toward which current work is directed.

| | |
|---|---|
| Oct. 1960–June 1961 | Pilot plant testing of 20 foods and pilot production of 10 foods. |
| July 1961–June 1963 | Production of 4 foods for military supply system. |
| 1962 | Larger role assumed by industry. |
| 1963 | Economic analysis for 10 foods completed. Procurement of a few selected items attempted by the military. |

The caution attending this schedule is expressed in words by the Army in a recent publication. In response to its own question, "Are irradiated foods acceptable?", the Army authors reply:

"So far, most foods undergo undesirable taste, color and/or texture changes at sterilizing doses. Generalization of this observation should not be misconstrued as dooming the commercial potentialities of the process. What is more telling at this early stage of research is that there are at least several commodities of commercial significance which show promise. Pork, bacon, pork sausage, fresh and cooked chicken, squab, shrimp, oysters, and green beans have all responded surprisingly well at sterilizing doses. At lower dosages, the storage life of strawberries and grapes has been prolonged. Mold growth has been suppressed for lemons in cold storage. Flour has been disinfested of insects by irradiation without significant alteration in its baking characteristics. Foodborne pathogenic parasites have been destroyed. In these cases, no undesirable changes resulted from the irradiation".

*4. Institutions engaged in research on irradiation of foods*

At the present time 80 institutions are engaged in research on irradiated foods under 100 contracts let by the U. S. Army. These embrace government laboratories, private research laboratories, universities, and industrial firms. The subjects of investigation range over the field; for example, can enamels for irradiated foods; control of meat enzymes; effects of irradiation on onions and potatoes; treatment of meat products to increase acceptability; pasteurization by radiation of cooked foods; radiation preservation of fish and fishery products; long-term feeding studies; carcinogenic studies; wholesomeness. In addition to the government-supported studies at the institutions named just below, there are, of course, many studies privately undertaken especially by the interested industries. The institutions marked by an asterisk are engaged in work under more than one contract.

Alabama Polytechnic Institute
American Can Company*
American Cyanamid Company
American Meat Institute Foundation
American Motors Corporation
Argonne Cancer Research Hospital
Armour and Company*
California Packing Corporation
Carnation Company
Continental Can Company, Inc.*
Cornell University*
Crown Zellerbach Corporation
De Bell and Richardson, Inc.
The Denver and Rio Grande Western Railroad Company
Doughnut Corporation of America

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Ekco-Alcoa Containers, Inc.
Florida State University
Food Research Laboratories, Inc.
General Electric Company
General Mill, Inc., Research Laboratories
Georgia Coastal Plain Experiment Station
Glutamate Manufacturers' Technical Committee
Hazelton Laboratories, Inc.
H. J. Heinz Company
George A. Hormel & Company
Industrial Bio-Test Laboratories, Inc.
International Salt Company
The John-Hopkins University
Kwik Kafe Coffee Processors of America, Inc.
Laboratory of Vitamin Technology
Long Island Vegetable Research Farm
Massachusetts Institute of Technology*
Medical College of Virginia*
Michigan State University*
Monsanto Chemical Company, Plastics Division
John Morell & Company
McCormick & Company, Inc.
National Canners Association (Western Research Laboratory)
National Canners Association
National Dairy Products Corporation
Nuclear Science and Engineering Corporation*
Ohio State University
Oklahoma State University
Oregon State College*
Pillsbury Mills, Inc.
Purdue University
Quaker Oats Company, Research Laboratories
Rath Packing Company
Reliable Packing Company
Reynolds Metals Company
Southern Research Institute
Southwest Research Institute
St. Louis University
Stanford Research Institute*
Stokely Van Camp, Inc.
Sunkist Growers
Swift & Company, Research Department
Syracuse University Research Institute
Texas A & M College
University of California, Davis
University of California (Los Angeles)*
University of Colorado
University of Illinois (Urbana)
University of Illinois, College of Medicine
University of Illinois School of Veterinary Medicine
University of Kentucky
University of Massachusetts
University of Miami School of Medicine*
University of Michigan*
University of Minnesota, (Hormel Institute)
University of Pennsylvania
University of Texas
U. S. Department of Agriculture, Plant Disease Laboratory
U. S. Dept. of Interior, Fish and Wildlife Service
Utah State University
Vanderbilt University School of Medicine
Virginia Polytechnic Institute
West Virginia Pulp and Paper Company
Western Utilization and Development Division, U.S. Department of Agriculture
Wisconsin Alumni Research Foundation*

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# 6. THE POTENTIALITIES OF FOOD IRRADIATION IN EUROPEAN COUNTRIES

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# The Potentialities of Food Irradiation for European Countries: Animal Products

H. R. BARNELL
Ministry of Agriculture, Fisheries and Food, Great Westminster House, Horseferry Road, London S.W.1

It is suggested that the first development of a trade in irradiation meat and meat products in Europe may be in "pasteurized" meat and that this trade might expand with increasing refrigeration facilities.

Later, should radiation sterilization of meat become a practical proposition, and if the present trends in imports of meat give a true indication of a general desire for more meat and meat products, there might be a large demand for the radiation sterilized product provided the quality were good and the price right in relation to the purchasing power of the peoples. Such a development might well cause some displacement of existing methods of preservation.

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## INTRODUCTION

THE main uses of ionizing radiations which appear to have possible applications to foods either in the near or the more distant future may be classified as:

(1) disinfestation of foods in bulk or in packaged forms
(2) prolongation of dormancy in resting organs, e.g. potatoes, onions
(3) inactivation of specific undesirable organisms in foods, e.g. *Trichinella spiralis* in pork, *Salmonellae* in frozen or dried egg
(4) "pasteurization", i.e. the inactivation of bacteria etc., in the vegetative form but not in the form of resting spores
(5) radiation preservation, i.e. preservation by complete sterilization.

### (3) *Inactivation of specific organisms*

In connexion with animal products interest lies mainly with "pasteurization" and sterilization treatments, but brief notice should be taken of the inactivation of specific organisms in pork and in egg products. Doses as low as 25,000 rads are sufficient to remove the danger of pork causing *Trichinosis* without giving rise to radiation odours or flavours in the meat.

*Salmonellae* in egg products may cause food poisoning. They are inactivated by doses of 0·3–0·5 Mrad with negligible effects on the baking quality of the products.

These treatments may be regarded as sufficiently advanced to be now technically feasible if required. *Trichinosis* is not a major problem in the United Kingdom when pork products are normally well cooked, but it is serious in some countries. *Salmonellae* in egg products present a continuing problem everywhere these products are used and a simple cheap method of removing the danger is needed.

### (4) *Pasteurization*

As previously indicated, future large scale developments in the radiation treatment of meat and meat products must be in the direction of "pasteurization" and full sterilization. For pasteurization, doses of the order of 0·5–1 Mrad are involved while full sterilization requires doses of 2 Mrads or more.

"Pasteurizing" treatments extend the shelf life of some refrigerated meats and meat products by a factor of five. This extension of the shelf life would markedly diminish distribution problems where refrigeration is

available but it should be noted that pasteurized meat products should not be stored at room temperatures for more than very short periods.

In Europe the use of refrigeration for food is increasing and it is at least possible that the combination of "pasteurization" by irradiation and the development of refrigeration could have a synergistic effect. The development of each could be mutually advantageous. However, there are problems to be overcome before "pasteurization" of meat can be regarded as technologically satisfactory. The dose levels required even for inactivation of vegetative bacteria produce irradiation odours, especially in raw meat. In prepared meats these odours are less evident and may even be absent. On cooking the meat, the odours are greatly reduced and any flavour changes tend to disappear. In the case of many prepared meat products any flavour changes which may occur are masked by the highly flavoured additives present.

The "pasteurization" process may be regarded as near the level of technological development required for consideration of commercial exploitation.

(5) *Radiation sterilization*

The ultimate objective of irradiation treatments is the attainment of the stable product preserved indefinitely in its fresh or "natural" state. This implies a completely sterile product with all katabolic processes inhibited. The product should be capable of storage and distribution without requiring any control of its environment apart from that provided by its bacterial, gas, and vapour-proof container.

Direct treatment with doses of 2 Mrads or more, required to produce sterilization, generally has adverse effects on the meat or meat product, though the extent of these deleterious effects varies from product to product. Unpleasant odours and flavours may develop, changes in colour including bleaching of fats may occur and the texture of the product may be affected. Processed meats generally are less affected than raw meats and highly flavoured products such as various types of sausages may be acceptable. There is some apparent difference of opinion between British and American workers on what constitutes "acceptability" in some products. The difference appears to be due in part to the weight given to the offensiveness of the irradiation odour of the product before cooking. Many products which might provoke distrust before cooking are, apparently, quite acceptable when the odour has been dispelled during cooking.

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## NEED FOR ACCEPTABLE QUALITY IN IRRADIATED FOODS BEFORE THE PROCESS IS EXPLOITED COMMERCIALLY

It is evident that the successful development of these processes could revolutionize methods of food preservation and lead to the establishment of a new industry which would be of great benefit to consumers by providing them with new kinds of sound and wholesome foods, preserved essentially in their natural states. However, it is equally evident that there are still formidable problems to be solved before attempts at commercial exploitation are likely to be successful.

Nevertheless the prospects appear sufficiently important to justify much effort in terms of men and money to achieve the advances in knowledge required to attain the quality of product necessary for successful commercial exploitation.

The major problems are those of cost of treatment and quality of the processed foods. With the production of radioactive isotopes as by-products of nuclear power stations, and the possibility of combining in one reactor power production and food irradiation, the cost problem should not be insuperable. However, until the products are fully acceptable there can be no prospect of commercial exploitation in Europe. It does not necessarily follow that the products must be unchanged by treatment, but it does mean that a solution must be found to the

quality defects which are a feature of many of the present experimental products. These defects are not necessarily inevitable since methods of reducing or eliminating some of them are already known and new methods might be found. Present methods include irradiating in the frozen state or in an inert atmosphere, in the presence of free radical acceptors, combining irradiation with other methods of preservation, e.g. heat treatments and stimulating spore germination chemically so that reduced radiation doses are effective.

These quality problems present a challenge to research workers and should be regarded in this light rather than listed as arguments against intensifying the efforts to develop a revolutionary method of food preservation. Few countries can afford to ignore the potentialities of developing an economical method of preserving meat which would permit easy cheap distribution and enable the demands of their populations for meat to be more adequately satisfied.

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*Health aspects of irradiated meats*

In addition to resolving the problems of quality and acceptability it will need to be established beyond all reasonable doubt that the nutritional values of irradiated meats remain adequate and that their ingestion does not present any health hazard. Much work will be required on this and some is now in progress in the United Kingdom. A large programme of work has been and is being carried out in the U.S.A. and so far has shown that both animal and human tests have given favourable results. Losses of nutrients are no greater and may even be less than those that occur with other methods of preservation. Carcinogens do not appear to be developed and generally no effect on health has been observed in human volunteers in the U.S.A. who have eaten appreciable quantities of irradiated foods over short periods of time.

## ESTABLISHMENT OF AN INDUSTRY IN IRRADIATED MEATS

Caution is necessary in considering the speed at which a new industry in meat irradiation in Europe may be expected to be established. People, and perhaps particularly Europeans, are notoriously conservative in their food habits and changes, however desirable, take a long time to occur. Even though solutions to the fundamental and technological problems of food irradiation may be found in the next few years and cheap sources of irradiation may become freely available, it will still require time and money to convince the potential consumer that the new products are desirable. A personal opinion is that assuming that much greater resources for research and development are made available and that the present problems are not intractable, something like ten years will be required to establish a food irradiation industry in the United Kingdom. Possibly this time could be shorter in countries whose peoples have less conservative food habits than the British.

*Scope for a meat products radiation industry in Europe*

Any attempt to assess the size which an irradiated meat and meat products industry in Europe might attain must be extremely hazardous even when it is assumed that the basic and technological problems have been solved and that the processes have become economically feasible.

According to the FAO Yearbook of Statistics (1957), the total meat including prepared and canned meat exported in the world in 1956 was 2,760,000 metric tons, while Europe imported 1,210,000 metric tons. These figures include fresh, chilled and frozen meats as well as meat products preserved by other methods. They are quoted to show the present level of world trade in meat of which new methods of processing might expect to assume a share in the future.

In recent years there has been a trend in the direction of increased imports into Europe of most kinds of meat and meat

products preserved by refrigeration, canning, salting, etc. and a similar trend can be observed for certain dairy products, e.g. dried milk, eggs and particularly cheese.

These trends suggest that if irradiation treatments provide cheap and attractive methods of distributing meat and related foods of good quality, then there will be consumers to buy the products.

It is at least interesting to attempt to calculate how much more meat might be consumed in Europe if meat consumption per head of population were as high as in the U.S.A. (i.e. 95 kg per annum). The figures given in the FAO Yearbook (1957) for *per caput* consumption of meat in each country have been multiplied by the population figures for the countries taken from the same source. On this basis it is estimated that the present consumption in Europe is nearly 14 million metric tons of meat per year. At the U.S.A. level of *per caput* consumption, the figure would be about 27 million metric tons. It may be suggested therefore that there is a potential gap of 13 million metric tons to be filled if suitably priced irradiated meat can be supplied providing, of course, that the populations are ready and able to change their food habits in the direction of eating more meat. This figure gives a probably maximal indication of the extent of an expanded market for meat in Europe.

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# Intérêt de l'Irradiation des Fruits et Légumes pour les Pays Européens

PIERRE VIDAL
Fondateur-Gérant de "Conservatome" 18 Rue Seguin, Lyon (Rhône) France

L'auteur considère que l'irradiation est une méthode intéressante pour l'inhibition de la germination des pommes de terre et de tous les bulbes et tubercules, car elle est particulièrement efficace et ne nécessite que des doses peu élevées, de l'ordre de 6.500 à 10.000 rads.

Ceci peut-être d'une importance considérable en Europe, où les dégâts dûs à la germination peuvent atteindre de 14 à 19 pour cent sur une production totale de 120 millions de tonnes en moyenne.

Des résultats encourageants ont été obtenus pour les fruits et légumes. L'irradiation peut être intéressante commercialement pour augmenter la durée de conservation à l'état frais des fruits et légumes les plus périssables.

Elle constitue, d'autre part, une méthode très efficace de destruction ou de stérilisation des insectes et larves parasitaires qui détériorent diverses denrées d'origine végétale ou animale (graines de céréales, porcs, etc.).

La méthode de conservation des aliments par irradiation présente, d'une façon générale, de multiples avantages tels que d'améliorer la commercialisation des produits, d'assainir dans certains pays la chaîne de distribution, d'augmenter les variétés de produits offerts aux consommateurs, d'augmenter la qualité des produits mis sur le marché, de diminuer le coût de certaines opérations et, finalement les prix des produits.

The author considered that radiation presents an attractive method for the suppression of sprouting in potatoes and root crops because it is particularly effective in this respect and only low doses of 6500 to 10,000 rads were needed.

This might be of considerable industrial importance in Europe when the annual production of potatoes is about 120 million tons.

Encouraging results had been obtained with some fruits and vegetables and radiation might yet prove to be commercially attractive for prolonging the life of the more perishable fruits and vegetables.

When it is further developed, the radiation preservation of foods may well have some distinct advantages such as decreasing the loss during storage of potatoes and root crops, increasing the range of products entering commerce, lowering the costs of storage and making the distribution of some foods easier.

Vous avez entendu au cours de la séance d'hier les très intéressantes communications qui vous ont été faites concernant l'application des radiations ionisantes aux divers produits d'origine végétale.

(1) par M. BURTON du Royaume Uni et M. MIKAELSEN de Norvège pour les pommes de terre et les carottes
(2) par M. CLARKE du Royaume Uni pour les fruits frais et les légumes
(3) par Dr. KRAYBILL et Dr. CORNWELL pour la destruction des insectes dans les graines
(4) par M. TRIPP pour les fruits et légumes.

## PREMIERE PARTIE

Il résulte de ces travaux et de ceux effectués dans d'autres pays que d'importants progrès peuvent, d'ores et déjà, être enregistrés dans ces différents domaines en ce qui concerne tant les techniques d'irradiation et des traitements que l'étude des effets par l'analyse des résultats.

Je ne reprendrai pas ce qui a été dit par les différents orateurs; je me bornerai à prendre acte de leurs conclusions pour m'associer aux espoirs qu'ils ont exprimés et qui ouvrent des perspectives plus immédiates et nettement plus encourageantes que celles concernant certaines denrées d'origine animale.

Il faut tenir compte pour l'Europe de ce que l'intérêt de l'irradiation diffère pour chaque pays en fonction de leurs problèmes économiques.

*Pommes de terre*

Il paraît acquis que l'irradiation des tubercules de pommes de terre en vue d'une inhibition de la germination est techniquement possible, qu'elle surclasse en efficacité toutes les méthodes de dégermination connues, qu'elle peut être effectuée avec des doses faibles s'étalant d'environ 6500/10,000 rads suivant la durée de l'effet recherché, qu'elle est exclusive d'effets nocifs et moyennant des prix de revient qui sont acceptables. Ceci s'applique également à d'autres tubercules et bulbes et notamment aux oignons et aulx.

*Fruits et légumes*

M. CLARKE nous a cité les résultats favorables obtenus du point de vue de l'allongement des durées de conservation à l'état frais de divers fruits tels que pommes, prunes, fraises, pêches, tomates, citrons, oranges, bananes, raisins et de divers légumes, grâce à des doses d'irradiation acceptables.

*Désinsectisation des graines*

Vous avez entendu également ce qui vous a été exposé quant aux méthodes de destruction des insectes parasitaires de divers végétaux et notamment de graines de céréales, par l'irradiation directe des produits infectés, à des doses relativement peu élevées.

Les problèmes qui se posaient quant aux techniques d'emploi de ces méthodes et quant aux contacts de leur action apparaissent en grande partie résolus pour certains produits.

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## DEUXIEME PARTIE

Il est permis d'essayer dès maintenant d'effectuer une évaluation, en termes économiques, des perspectives qui vont s'ouvrir pour le continent européen dans un avenir qui n'est désormais plus très lointain.

Dans le cas de la *pomme de terre*, l'enjeu peut s'exprimer par quelques chiffres:

L'Europe est le plus important producteur de pommes de terre et le plus important consommateur.

L'Europe intervient pour une production moyenne de 120.000.000 de tonnes dont 15.000.000 pour la France. Il est admis pour certains que la perte totale du fait de diverses causes se situe entre 14 et 19 pour cent de ces chiffres, dont les 3/4 seraient dûs, directement ou non, à la germination. Si on ne retient qu'un chiffre de 10 pour cent pour les pertes dues à la germination, ces dernières représentent l'équivalent d'une valeur qui est de l'ordre de 300 milliards de francs français par an pour l'Europe.

On sait, d'autre part, qu'indépendamment des pertes dues à la dégermination, les difficultés d'effectuer les soudures d'une récolte à l'autre avec des produits intacts oblige, du fait des exigences du ravitaillement, à des transports de stocks d'une région à l'autre. Leur coût aggrave encore considérablement les chiffres de pertes énoncés plus haut, sans compter le prix élevé de ces produits du fait de leur faible abondance.

La production des *oignons* et *aulx* à l'échelle d'un pays comme la France représente, bon an mal an, 160.000 tonnes pour les oignons et 30.000 tonnes pour les aulx, qui se

déprécient notablement en valeur et en qualité pour les mêmes causes. L'arrêt de la germination que l'on pourra obtenir par l'irradiation à faibles doses, dans des conditions économiques représentent donc un gain très appréciable. Celui-ci existe dans des limites relativement moindres pour les oignons qui offrent, comme on le sait, une gamme de variétés assurant un approvisionnement plus continu.

Pour ce qui est des *fruits* et *légumes*, nul n'ignore combien la plupart d'entre eux sont sujets à une rapide détérioration, principalement ceux qui représentent les variétés les plus appréciées et les plus chères sur les marchés. Une prolongation de quelques semaines peut représenter de considérables économies en tonnage et en valeur.

L'irradiation à des doses de 0,15 à 0,2 Mrad jointe à la réfrigération à $+4°/+6°C$ permet de prolonger dans d'appréciables proportions la durée de conservation de nombreux fruits frais très périssables. Aux résultats donnés hier, il convient d'ajouter les abricots et les pêches dont la conservation, dans les conditions ci-dessus, est grandement augmentée.

A la lumière des résultats obtenus dans des études encore en cours sur les tomates, la durée de conservation de ce produit à l'état frais pourra être prolongée dans de notables proportions.

En ce qui concerne les légumes feuillus, les résultats obtenus sont encore assez divergents et il reste beaucoup à faire dans ce domaine avant que l'on puisse faire un pronostic d'avenir et chiffrer des espoirs.

Pour les légumes et les fruits, qui sont produits principalement pendant l'été, l'étalement de l'approvisionnement des marchés sur plusieurs semaines supplémentaires permettra de limiter les déperditions, du fait de la chaleur, et celles résultant parfois de surproductions saisonnières ou locales, pour le plus grand profit des producteurs et des consommateurs.

Ces résultats seront parfois obtenus par l'utilisation courante de la réfrigération et de l'irradiation.

Pour ce qui est des céréales, la limitation des dégats d'insectes qui pourra être réalisée, comme vous l'a démontré dans son excellent travail, M. Cornwell, et son équipe de Wantage, est de nature à engendrer elle aussi des gains portant sur des tonnages et des valeurs d'un ordre de grandeur très élevé.

On a avancé pour des pertes sur les blés, du fait de leur infestation lors des manipulations, le chiffre de 3 pour cent. On voit que le gain d'un seul point sur ce pourcentage appliqué aux très gros tonnages qui représentent les productions des pays consommateurs de pain, ce qui est le cas de tous les pays d'Europe, peut correspondre à des sommes très élevées. Par exemple, le gain d'un point représenterait pour la France une économie de près de 2 milliards de francs.

## TROISIEME PARTIE

Les incidences économiques que peut avoir dans l'avenir le développement de l'irradiation de certains produits alimentaires apparaissent, d'ores et déjà, devoir être extrêmement favorables dans le cadre d'un pays déterminé.

Mais les chiffres qui les traduisent se multiplient, plus qu'ils ne s'additionnent si, élargissant les horizons, on considère le problème sur un plan s'étendant au delà des cadres nationaux, c'est-à-dire à la dimension des communautés et marchés englobant plusieurs pays, et plus encore si les communautés associent les économies des pays continentaux et de pays ou territoires d'outre mer.

La prolongation de la durée de stockage ou la conservation à l'état frais des produits pourra permettre d'envisager pour certains d'entre eux des débouchés plus lointains, d'ouvrir certaines régions à la consommation de produits nouveaux, de favoriser la circulation des produits, et d'étaler leur distribution, dans le temps et l'espace, au stade de la consommation.

Les charges des investissements ou

installation de stockage, en navires, wagons ou véhicules spéciaux pourront, dans certains cas, être réduites. Dans d'autres cas, il leur sera assuré une meilleure utilisation, donc baisser le coût de leur intervention.

Deux exemples, bien connus des exportateurs de fruits des pays du Sud de l'Europe, permettent d'illustrer certaines des possibilités nouvelles qu'il sera, on peut l'espérer, possible d'envisager lorsque les techniques seront au point et admises.

Les wagons de fruits qui sont expédiés au Sud de l'Italie, sur les marchés de Londres, n'arrivent à Covent Garden qu'après 6–8 jours depuis la cueillette, par la voie Chiasso–Dunkerque–Douvres, avec deux reglaçages des wagons réfrigérants en cours de transport. La longeur de ces délais implique la cueillette des fruits dans des conditions souvent défavorables, qui nuisent à leur qualité, à leur sélection, à leur conditionnement etc. . . .

Il en est de même de certains fruits expédiés d'Espagne, d'Italie du Sud, de la Grèce ou du Sud de la France vers les pays scandinaves. Les conditions de transport sont alors tellement difficiles, longues et souvent aléatoires, qu'elles rendent, dans la pratique, parfois impossible l'approvisionnement normal de ces pays pour certains fruits. L'allongement de la durée de conservation de ces produits par irradiation seule, ou par l'utilisation conjuguée de la réfrigération et de l'irradiation, permettra aux pays producteurs l'ouverture de ces nouveaux marchés lointains. L'augmentation de leur production qui en suivra sera d'un intérêt certain pour l'élévation du pouvoir d'achat d'une partie de la population des pays producteurs, tout en améliorant et en diversifiant l'alimentation des nouveaux pays consommateurs.

Ceci sera vrai également pour les régions productrices des pays tropicaux dont l'économie est en partie rattachés à celle de l'Europe.

On pourrait multiplier les exemples de ce qui sera, selon nous, possible dans un délai relativement proche pour un certain nombre de produits.

Les techniques nouvelles que les industries et commerces de l'alimentation européens peuvent espérer avoir bientôt à leur disposition, grâce à l'irradiation, doivent avoir pour résultats:

(1) d'augmenter les quantités de pommes de terre, oignons, fruits frais, céréales, mis à la disposition des populations grâce à la diminution des pertes sur stocks ou en cours de transport;
(2) d'améliorer la commercialisation des produits;
(3) d'assainir, dans certains pays, la chaîne de distribution;
(4) d'augmenter les variétés de produits offerts aux consommateurs;
(5) d'augmenter la qualité des produits mis sur le marché;
(6) de diminuter le coût de certaines opérations et finalement le prix du produit.

Tous ces résultats ne pourront qu'être profitables à l'amélioration du niveau de vie de l'Europe et c'est pour cette raison que nous pensons qu'il convient d'intensifier les recherches pour permettre d'utiliser les résultats dès que possible avec des techniques offrant toutes garanties.

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# Radiation in Relation to, or in Combination with, Other Processes

M. INGRAM
Low Temperature Station for Research in Biochemistry and Biophysics,
Department of Scientific and Industrial Research, Cambridge

For any application, three aspects must be considered: technical feasibility, suitability for the industry concerned, and competition with alternative procedures.

Sterilization by radiation alone is rarely feasible, because of quality deterioration. Preliminary irradiation however permits sterilization with less heating, a combination potentially useful in canning: though antibiotics with similar effect would be serious competitors.

Lower-dose "pasteurization" procedures can kill enough spoilage microbes to extend storage life of meats, fish and fruits, often without too much loss of quality. The proportionate increase in time to spoilage is greater at low temperatures. Storage below 5°C is also desirable with non-acid foods, to avoid possible development of food-poisoning bacteria likely to survive the irradiation. The requirement for refrigeration is a serious limitation. Apart from microbiological problems, the quality changes due to irradiation are appreciable and must be evaluated commercially before their importance can be assessed: at present, freezing gives meat of superior quality. A particularly interesting "pasteurization" process is the destruction of salmonellas in frozen foods, without thawing.

Certain low-dose treatments which appear feasible—to disinfest grain or inhibit potato sprouting—would cause such serious disorganization of the industry that fumigation processes seem more economic.

I HAVE been given the difficult task of summarizing certain aspects of the proceedings of this meeting after surveys by two previous speakers. As I had no knowledge of the subject matter of their talks until I heard them, to attempt to review the remnants of the proceedings is scarcely possible.

I therefore propose instead to give a purely personal assessment of the prospects for some aspects of radiation processing with which this meeting has been particularly concerned, mainly those where the target is microorganisms. It is sufficiently evident that the advantages of radiation processing could be great: the question is, whether any such process would in fact be successful in practice?

In forming an opinion about this, three things have to be considered. First, obviously, is the technical feasibility of the particular process. Second is its general suitability for application in the industry in question. Third is the existence of alternative, and possibly more suitable, procedures.

There can be no doubt now that irradiation treatments are in principle possible: it is abundantly clear that microbial spoilage can be delayed to a significant, and sometimes striking, extent. But it may still be doubted whether such a process would be technologically feasible; for it has yet to be convincingly shown that the products would be of satisfactory quality when judged by commercial criteria. This is indeed a difficult question, highly subjective, with different criteria in different countries, and at present impossible to investigate properly because of restrictions on the widespread consumption of irradiated food. Our experience with irradiated meats, even with low dose treatments, is that a decline in quality becomes obvious well before microbial spoilage terminates the shelf life of the

product; so that the irradiated is inferior to the frozen commodity. Manufacturers in this country indicate that they want any new process to give a product which is better than, or at least as good as, that processed by existing means. So far, irradiated meats have not met this requirement.

The introduction of irradiation would call for expensive equipment and personnel, and the techniques involved are considerably more complicated than those commonly used in the food industries. Because of this, some centralization of processing facilities is likely to be needed, which is plainly unsuited to an industry operating in small units over a wide area. For instance we have already heard that, with grain, the transport costs of the necessary reorganization would be prohibitive; and that, because of its greater flexibility in application, a fumigation process seems much more suitable. Similarly with potatoes: without doubt sprouting can be prevented with radiation, though with changes in quality whose significance is undecided; but here too it appears that a method better suited to the industry's organization is available, in fumigation with nonyl alcohol, for which reason we have ceased to irradiate potatoes.

With such considerations in mind, let us now look at some of the antimicrobial processes which have already been discussed at this Conference, beginning with those aiming at sterility.

The outlook for straightforward sterilization procedures is frankly discouraging. In several years of intensive search, no combination of processes has been discovered which offers any real hope of reducing appreciably the large radiation doses needed to achieve a satisfying degree of sterility; doses which, with rare exceptions, yield products which we regard as commercially unacceptable. In addition, there would be severe requirements for the containers: the use of plastic films still leaves much to be desired, and at present only metal boxes fulfil the requirements. My impression of sterilization processes is, then, that they are at present not technically feasible, except perhaps with a few foods in U.S.A.

The combination of radiation with heat processing affords a more encouraging prospect. One can clearly expect to use less heat for sterilization. The canning factories appear suitable to accommodate and use radiation facilities. Further study is required to determine whether bacterial spores remain inactive after irradiation and heating in the same way that they do after heating alone; and, because such investigations can only be made by large canning research organizations, I can hardly form any experienced opinion about them. It must however be borne in mind that the use of polypeptide antibiotics would clearly be simpler and probably a serious competitor to the combination of radiation with heat processing.

When we come to consider the pasteurization type of procedure, i.e. use of low doses insufficient to sterilize, it is important to realise that storage below 5°C is desirable for the non-acid perishable foods, like meat, fish, eggs etc. which form our chief interest. Otherwise there is an apparent possibility that pathogenic organisms might develop. Experience with chickens indicates that the use of radiation, especially with antibiotic treatment, gives a very useful increase in storage life under refrigeration, though progressive loss of quality reduces the advantage gained to an appreciable extent. In the case of beef, a useful increase in microbiological storage life is apparently obtained only with a dose which considerably enhances the prospect of the meat fat becoming rancid during storage. Conflicting results have been obtained with sausages and precooked meats. In the case of fish a careful study needs to be made of the quality of the product in storage after low doses of radiation.

The combination of low temperature storage with irradiation appears also to be beneficial in the case of fruits and vegetables; though there is no pathogenic hazard here, the beneficial effect of irradiation is greater the lower the temperature of storage. But here, again, careful study needs to be made of the quality of the product, in particular commercial evaluation seems desirable.

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While the prospect for pasteurization processes is hopeful, I should hesitate to recommend any application until the quality changes have been evaluated more definitely. And, even were technical feasibility successfully achieved, the firms organizationally capable of using the process and exploiting its marginal advantages are few, at any rate in Britain.

The inactivation of pathogens by radiation pasteurization is an attractive idea, but the number of actual applications is limited. The killing of *Salmonellas* appears to be the most promising such process. *Salmonellas* can be eliminated from dried egg by heating or treatment with ammonia, or from liquid egg pulp by heat pasteurization during manufacture; but once the latter material has been frozen, radiation affords a unique method of eliminating *Salmonellas* without opening the container or thawing, and without serious loss of quality. Radiation might conceivably be useful similarly to destroy *Salmonellas* in frozen meat, but the dose probably required (0·5 Mrad) might cause some adverse quality changes, especially with beef. Although either process would seem useful, their application in Europe would require considerable organization in transport of the infected material through treatment centres, and it is not yet clear whether this would be practicable.

In summary therefore, except for the rather unusual case of inactivating *Salmonellas* in frozen egg, pasteurization of meats—especially chicken—seems to me the most immediately promising application of radiation in food preservation and, accordingly, these are the processes which my group is investigating. Let me remind you again that such processes, though coupled with antibiotics, require refrigeration. Even in Britain, a comparatively highly-developed country, our industrial distribution chains could not at present meet a wide necessity to maintain temperatures below 5°C, and domestically, the use of refrigerators would have to be extended considerably. Moreover, granted these facilities, past experience with canned hams indicates plainly that the precautions would often be ignored; while the consequences of such neglect might be far more dangerous than with canned hams, and would require special consideration. Even if we had a radiation pasteurization process which was really effective yielding a product entirely satisfactory, and an industry able and willing to use it, there might still be serious administrative problems in establishing a process of this kind, which is clearly unsuitable for under-developed countries.

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# Discussion

Dr. Mossel (The Netherlands): Could we not arouse more interest in the general adoption of refrigeration in view of its absolute necessity if radiation pasteurization is to be used at all?

Mr. Oxley (U.K.): The disinfestation of grain using radiation seems superficially promising but it must be remembered that even if the radiation were free, transport costs would be more expensive than current methods of disinfestation. Some form of technological revolution would be needed to implement radiation processing. Another disadvantage is the necessity to work in large units, since the highest losses of grain occur in countries such as India where material is consumed on the spot. Small radiation units might have application in this situation. Fumigation and other techniques must always be very much cheaper than radiation in the existing circumstances of the European grain trade. The biggest losses in grain are caused by insects in the hotter more backward countries—India, Africa, South East Asia, etc., where grain seldom moves more than a few miles. This requires much smaller irradiation units than have so far been proposed. Effort should be made to make small simple irradiation units which can be widely distributed, then the irradiation technique for insect destruction in grain will come into its own.

Dr. Betty Hobbs (U.K.): In reply to the comment by Dr. Mossel, the salmonella contamination of egg products probably arises from eggs laid by hens showing no symptoms of salmonella infection. Until this source can be controlled there are only two methods of destroying the salmonellae:

(a) by pasteurization of the liquid mix or heat treatment of the dried product e.g. flaked albumen or
(b) by irradiation of the liquid, frozen or dried material.

The incidence of food-poisoning from contaminated egg products is indeed a very real problem.

I would like to ask Dr. Ingram, in view of the frequent occurrence of *Clostridium perfringens* food poisoning from pre-cooked boiled chicken, and also of occasional outbreaks of salmonella food poisoning from poultry cooked in various ways, if work has been carried out on the proportion of poultry carcasses contaminated with heat-resistant poisoning strains of *Cl. perfringens* and with salmonella, and what is the effect of irradiation of the carcass on these organisms?

Dr. Ingram (U.K.): In reply to Dr. Mossel, it would certainly be better to eliminate salmonella earlier, preferably by measures designed to prevent infection of eggs as laid, or failing this, by pasteurization of the pulp at the factory before freezing. Unfortunately, we cannot demand these changes in foreign countries, and have to deal with the situation as it is.

In reply to Dr. Hobbs, the doses envisaged, 0·5–0·75 Mr, should kill the salmonellas, but certainly not eliminate the spores of *Cl. perfringens* and other anaerobes. However, we never observe that *Cl. perfringens* takes part in the normal spoilage of chickens and, especially, temperatures below 5°C, as proposed, would protect against it.

Dr. Mossel (Netherlands): Would it not be much more effective to insist on killing the salmonellae by appropriate heat pasteurization of the egg pulp prior to packaging, and freezing, rather than to go on accepting salmonella contaminated products and the need for radio pasteurizing of these after it has been packed in tins?

Dr. Hobbs (U.K.): I should like to ask Dr. Ingram whether the pathogenic organisms in pulped egg include *Clostridium welchii* and salmonellae.

Dr. Ingram (U.K.): Salmonellas can,



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of course be killed by pasteurization procedures. *Clostridium welchii* cannot be killed by pasteurization procedures but spoilage of chicken does not involve *Clostridium welchii* from the evidence which is available. In answer to Dr. Mossel's point, heat pasteurization is a proven process, but when the egg arrives in the frozen condition it may well be more acceptable to irradiate it. Since we cannot influence the egg producers to pasteurize the eggs at the source we have to do it in this country in the condition in which it arrives.

Mr. JEFFERSON (U.K.): Electrical machines and radioisotopes are much more complementary than competitive. The machines are at their best in dealing with small packages and radioisotopes are pre-eminent for treating large packages and boxed material.

Dr. HANNAN (U.K.): I would like to support Mr. Jefferson's remark completely. The two types of equipment are not competitive; the only difficulty arises when one has to choose between one or the other. Preferably one should have both.

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# Final Comments

R. S. HANNAN
Research Department, The Metal Box Co. Ltd., Kendal Avenue, Acton, London

VARIOUS potential uses for radiation processing of foods have been discussed in great technical detail and it remains for the delegations to reflect upon their significance. Reflection may well be lengthy since the implications will vary widely from country to country and this paper is mainly intended as a first contribution to such thought. Many factors are relevant to the interests of individual countries including, for example, the nature of their foodstuffs and the general status of their food and atomic technology, and the speaker can claim no detailed knowledge on these points. No attempt has been made, therefore, to prepare a comprehensive review of all major aspects of the subject, but rather a few topics have been selected for comment on a purely personal basis.

The first point which must be made is that the display of the subject which has been achieved by the Meeting has been distinguished by its breadth and quality and this must surely have been the most complete such display yet seen in Europe. Comparable meetings have, of course, been held on various occasions in the United States of America and the presence of leading workers from that country has provided a valuable link with the spirit of those meetings, in addition to the stimulus of their personal contributions to our discussions.

The Meeting has also been noteworthy for its objectivity of tone. It was pointed out at the commencement that the Wageningen Conference had recognized that nothing was to be gained in this field by over-enthusiasm, but it must be equally clear that nothing is to be gained by under-enthusiasm and one of the difficulties, particularly for the non-specialist, is to strike a suitable balance. The comprehensive survey of the scientific background of the subject here presented should permit a realistic appraisal of the present known facts and an appreciation of those aspects which are still speculative, and this in its turn should form a basis for the prosecution of meaningful research. It is submitted that the present technical facts are such as to justify proceeding with the work in a mood of *cautious optimism*. Inherent in this approach should be recognition that there is at least a possibility that any particular line of work may fail to lead to direct practical application but, as in any other branch of advancing technology, each individual will assess the degree of this possibility differently and must be prepared to change his assessment with time and circumstance.

Turning to the detailed aspects of our discussions many points of interest have been presented, among which a few may be mentioned specifically.

In the first place it has emerged quite clearly that there has been no enthusiasm at this Meeting for the application of doses greater than about 1 Mrad except in very special cases.

Secondly there has been a much needed discussion of various public health aspects of the subject.

(1) The wholesomeness of irradiated foods has been discussed at length including the food legislation aspects, resulting in valuable guidance on this particularly complex issue.

(2) It has been pointed out that ionizing radiations might be used for controlling harmful organisms in food, such as *Salmonellae* and various human parasites.

(3) There has been a useful ventilation of the possibilities that food poisoning organisms might grow in irradiated

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foods during storage. This requires particular attention since, apart from the basic undesirability of food poisoning *per se*, any such occurrence could be expected to result in loss of consumer confidence in all types of irradiated foods.

In so far as new technical developments leading to widening spheres of possible application are concerned, it must be recognized that this Meeting has produced few reports which fall into that category and the papers presented have, in fact, mainly been concerned with amassing essential detail relevant to techniques which have been known for a year or two at least. This is a perfectly natural state of affairs which suggests that radiation research is now coming into line with other advances in food technology. In similar manner there is apparent duplication of work in different laboratories and different countries. Such studies would be carried out in any other field even for the mere training of staff and gaining experience, but in this field there is a special point in such investigations since the food products of different nations may respond differently to irradiation. We have seen, for example, that there appears to be a difference in radiation response between British and American bacon. Individual national reactions to flavour change may also vary; some food products may even be peculiar to individual countries; and microbiological problems would certainly vary from country to country. There is, therefore, everything to be said for investigational work, if only of a screening nature, to be carried out on a wide basis.

It is only natural that an important part of our discussion should have been concerned with the various available methods of irradiation and the economics of their use. These discussions have been so complete that specific mention is not required at this point except that as far as facilities for research, rather than production, are concerned it may be noted that, in the experience of the speaker, radioactive sources have the advantage of uniform and reproducible dose rate whereas electron accelerators have the advantage of widely variable dose rate. The latter is one of the variables which has still to be investigated thoroughly. Such facts as well as questions of cost and irradiation capacity should be considered in planning for any experimental facilities.

Perhaps the most immediate question which arises from our deliberations is whether commercial development of other methods of food treatment should be modified or abandoned as a result of developments in the field of irradiation. It is submitted that there has been no indication in this Meeting that radiation processing is likely to displace any conventional technique at least in the immediate future, and it should therefore be thought of only as supplementing existing techniques rather than displacing them. In the case of refrigeration we have even seen that the development of radiation pasteurizing would be expected to create a greater demand for chilled storage.

A further important question is whether special legislative provision is required concerning this process. The significance of this question varies widely from country to country and cannot be pursued here.

The main remaining questions concern the needs for further work in this field. Much of this falls under the heading of amplification of existing data and the following are suggested as warranting further study.

It is desirable to explore fully the significance of lower dose procedures requiring less than about 25,000 rads; for example for the control of human parasites, insect pests and vegetable sprouting.

More work should be carried out on the extension of storage life achievable under commercial conditions with perishable foods treated with moderate or low doses of radiation.

It would be desirable to define the effect of irradiation on important foodstuffs peculiar to individual nations, particularly to determine whether there are any unexpected differences.

For new work it will, as always, be necessary to rely chiefly on the natural inventiveness of the workers immediately

concerned with the research, but the main potentialities for discovery seem to be in the field of combination processes; that is the use of various methods of microbiological control, including radiation, in such a way that the total effect is greater than the sum of the individual effects. The number of possibilities is very great, but here it must be emphasized that irradiation work is not alone in its interest in this aspect of food processing. The combination of antibiotics with heat or with refrigeration, or the combination of various methods of packaging with a refrigeration are examples, and in this field we should reflect that it is the microbiologist who must lead the way. Microbiologists competent to work in such complex fields are, of course, limited in number and careful consideration should clearly be given before any proposals are accepted which may grossly overload these workers. The same consideration applies also, to a lesser degree, to trained food scientists in general.

Continuing work is also required on chemical, microbiological and wholesomeness studies to provide a background of basic experience. As an example of the type of information still needed, it would be valuable to have a basis for an objective method of determining whether and to what extent a foodstuff has been irradiated. This would permit control of the process and guard against the possibility of a product receiving more than one irradiation treatment. It would be natural to seek a chemical method for such a test but this would conflict with the parallel endeavour to carry out irradiation procedures in such a way that chemical change is kept to a minimum.

In prosecuting these and the many other studies which can be suggested, an essential requirement seems to be not to attempt to travel too fast. A time scale of 5 to 10 years has been suggested as representing the time before any appreciable commercial developments could be expected and this figure conforms with the speaker's experience. Considerable work will be involved and many false trails will be followed before the full potentialities will have been explored, but there can be no question of the value of this work, either in leading to direct practical application or in contributing to knowledge of food science and radiation science, and thereby to wider developments in the future.

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# 7. PROPOSALS FOR CO-OPERATION IN RESEARCH IN EUROPE

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# Co-operation in Research in Europe
## Introduction

J. KUPRIANOFF
Bundesforschungsanstalt für Lebensmittelfrischhaltung, Karlsruhe, Germany

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BEFORE asking Dr. Silow to present his statement about co-operation in research in Europe, I should like to make some introductory remarks concerning this particular subject. These remarks might be taken at the same time as an explanation for having a special place on the agenda for this subject since it might be argued that there is little to discuss about co-operation in research in this field.

I think the first problem we have to consider is whether there is a need for international or even intra-European co-operation in research in the field of the application of ionizing radiations to food preservation; secondly we may ask ourselves whether the time has arrived for establishing the necessary contacts and for supporting the idea of intensive international co-operation in this field; and thirdly we might consider the kind of co-operation which it would be most profitable to initiate, to support and maybe to assist financially.

Considering first the question of the need for co-operation in research, we should keep in mind the extremely high initial cost of equipment necessary for carrying out adequate research on food irradiation, the need for highly skilled and well trained research workers and auxiliary staff, and the high operational and maintenance costs of the laboratories and equipment concerned.

I suggest that these considerations make it worth while for this meeting to discuss possible forms of scientific co-operation in this field where co-operation seems to be even more important than in other fields of food research.

Results so far obtained in research laboratories in different countries indicate that this is a field of work with which research workers in other branches of food technology should maintain contact, and in which they might also engage in the not too distant future.

The field of work concerned with food irradiation is so extensive and the number of problems remaining to be solved are so great that co-operation would be very profitable even in countries such as the U.S.A. and the U.K. where already much research work on the subject is in progress.

For small countries wishing to start work in this field, co-operation would be even more valuable since thereby programmes of work could be carried out more economically and results would be obtained more quickly. It seems very likely that, in small countries, radiation research will not be started in several centres but that the work would be confined to one or two centres. Nevertheless there should be an exchange of opinions between scientists concerned with the same or similar problems. Moreover contacts should be established with specialists in other countries.

Next we might consider whether the time has arrived for such co-operation. While trying to judge the present situation as objectively as possible, I would suggest that the sooner that the first steps towards this objective are taken, the better will be the results. If this is not done, there will be the danger that the gap in scientific knowledge and experience in this field between scientists in the larger countries with advanced programmes of work and those in smaller countries just developing their programmes will become so great that adequate and promising co-operation between these two kinds of countries may become very difficult.

Moreover, it is probable that, depending on conditions in each individual country, different kinds of co-operation may be advisable at different stages of development of programmes of research in food irradiation.

As regards possible forms of international co-operation on scientific and technical problems, I should like to stress the fact that these could vary in several respects: the co-operation could be occasional and of short duration or it could be permanent; it might include only one small problem or be on a consultative basis and yet it might extend to intensive team work. Moreover the intensity may itself vary so much that it would be well to distinguish between co-operation and collaboration and both of these forms might involve individual scientists or groups of research workers.

Bearing these facts in mind, I suggest that international co-operation might take one of the following forms:

1. *Personal contacts between research workers*

These may begin by exchange of correspondence and of publications and lead to exchange of visits during which detailed personal discussions would occur. In my opinion, visits of this kind to special laboratories should be encouraged.

2. *Exchange of younger research workers*

This might well be effected by the granting of fellowships (say for 1 to 6 months) by the host or the home country. International organizations could render valuable help by giving advice and financial assistance.

3. *Co-operation between research institutes in particular programmes of research*

For co-operation of this kind to be successful it is essential that mutual confidence must exist between the institutes concerned and this can only be established by personal contacts. The institutes themselves would probably have to take the initiative to achieve co-operation of this kind, but governments could also help by encouraging such plans and by giving financial assistance. Particular subjects which might be appropriately studied in co-operative programmes of research are for example analytical methods and dosimetry.

4. *Participation of scientists from other countries in national scientific meetings or symposia*

This would be particularly useful for the younger and enthusiastic research workers and financial assistance from governmental sources or from international organizations would be most useful since it is often the case that the institutes themselves have not sufficient funds for travel of this kind.

5. *The organization of and participation in international meetings*

International meetings such as the present one and the UNESCO Conference on Isotopes held in the Fall of 1957 in Paris attended by representatives of professional and governmental organizations are a valuable method of co-operation.

6. *Exchange of information on research programmes*

This may raise rather delicate issues, but if such an exchange could be effected, say at the beginning and end of each year, it would help to achieve some form of co-ordination of effort. Moreover, the publication of statements describing research in progress—a course followed for instance by the International Institute of Refrigeration—could be very helpful.

7. Finally the *offering of some special services* to research workers in other institutes—e.g. the use of radiation facilities or measuring equipment, consulting services, etc.—would be very useful.

We shall probably find that within these various possible forms of co-operation, some will be more appropriate at the beginning of international co-operation while others will be more appropriate later. Moreover the form of co-operation may differ for different institutes and for different countries.

Whatever the case, it seems to be both reasonable and essential to have international co-operation in this special field.

I believe that FAO which has already

taken an active part by organizing the present meeting and which has considerable experience in the organization of international co-operation in various other fields can advise us well about the means to use to bring about international co-operation in the particular subject with which the present meeting is concerned.

By giving appropriate advice and by making specific recommendations, it might also help to obtain financial assistance from governments for the purpose of improving international co-operation in this field.

Dr. Silow is the main speaker on this subject and I am most grateful to him for undertaking the task of considering this important problem. I am sure that the meeting will be very interested to hear what he has to say, and I hope that, as a result of our discussions, we may find means for co-operation that are appropriate for all of us.

I call on Dr. Silow to address the meeting.

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# Co-operation in Research in Europe

R. A. SILOW
Food and Agriculture Organization of the United Nations, Rome, Italy

AFTER the Chairman's very comprehensive summary of the possibilities of co-operation in research in food irradiation for European countries, I shall speak only very briefly. In the course of this meeting it has become clear that a great deal of research is required in many aspects of food irradiation, and that there is a general recognition of the need for international co-operation in that research.

The possible types of co-operation range from an extremely close collaboration, perhaps through a centrally located and jointly financed research institute, to a very loose type of co-operation such as is represented by personal contacts between workers in different countries, or the exchange of information through meetings of the present type.

Clearly Europe is not yet ready for any really close collaboration in research in this particular field—that is something that can only grow from a demonstrated need for it. At present we should give consideration to the various forms of co-operation to which the Chairman of this session has referred.

There are various ways in which an international organization like FAO can be of assistance, and this Meeting should therefore give consideration to the action which it wishes FAO to take in the future.

FAO could, for example, in association with other international organizations working in this field, organize a small technical working group composed of technical representatives of countries actively engaged in research in this field, or planning to develop an active research programme. Such a committee could keep developments under continuing review, as has been done for the first time on the international plane by this meeting.

Under the aegis of such a central overall food irradiation committee there could be various technical working groups to consider more limited aspects of the general field. For example, the delegate of the Netherlands, speaking as Chairman this morning, emphasized that there is need for a common approach in Europe to the technical principles which must form the basis of legislative control of the use of irradiated food. A technical group could be established to deal with such matters, either under the aegis of the overall technical committee which has been mentioned, or within the framework of existing arrangements such as the FAO/WHO Expert Committee on Food Additives.

Another form of international co-operation which has been mentioned is the provision of fellowships for study in countries with experience of this new technique. Both FAO and the International Atomic Energy Agency are already awarding fellowships in this field. FAO, however, can make only limited funds available for this particular field, and the nature of the contribution towards international co-operation that FAO is best in a position to offer is to provide a continuing platform for contacts at the technical level, in meetings of this present type or in a more formally constituted committee such as has been mentioned. FAO looks forward to receiving some guidance from this meeting as to the ways in which it can best be of assistance to research workers in this field.

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# Co-operation in Research in Europe—Exchange of Information between Microbiologists Concerned with the Radiation Preservation of Food

B. A. BRIDGES

U.K.A.E.A., Isotope Research Division, Wantage Radiation Laboratory, Wantage, Berks

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DURING the discussion on microbial aspects of food irradiation, it was suggested by Professor Manil of Belgium that the microbiologists concerned with the radiation preservation of food should make arrangements to exchange information and ideas, since there was much which could be of great value but which could not be covered by the present discussion owing to the more general nature of the Harwell meeting. Dr. Mossel, Secretary of the Section on Food Microbiology and Hygiene of the International Association of Microbiological Societies, said that the Association was interested in this and his Section was prepared to co-ordinate such an exchange.

After the conference, enquiries were sent to all who had expressed interest, and as a result twenty-six scientists contributed information to a circular letter about their work, radiation sources, future plans and bibliography. It is hoped that the liaison which is now established between workers will be continued by them, and it is felt that the outcome may well be a much closer co-operation between workers engaged in similar projects.